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Cross-Country Urban and Rural Measurements of NO_x and SO_2

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The data were gathered from Denver, Colorado, to Raleigh, North Carolina, in August 1976, and from Los Angeles, California, to Raleigh, North Carolina, in November 1976. In November the laboratory stopped at six rural locations to record time-averaged data, which are also presented.

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CROSS-COUNTRY URBAN and RURAL MEASUREMENTS of NO_x and SO_2

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Total sulfur and oxides of nitrogen measurements, gathered along two long-distance routes across the United States, are presented in this report. Supportive information describing the instrumentation, procedures, moving laboratory, and regional meteorological conditions are provided.

Over 300,000 measurements were made. Points consisting of ten-measurement averages have been plotted; they are also presented in a graphic analysis. This analysis consists of frequency distribution plots of segments of the trip, for example, through rural areas and crossing urban developments. These plots provide a means of classification of the degree of pollution present and monitored with this moving laboratory technique.

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Data collection and equipment maintenance during the cross-country trips were performed by the persons from Environmental Measurements, Inc. and the U.S. Environmental Protection Agency listed in the table below.

Table 1

Participants in the Cross-Country Measurement Program

Individual	Trip 1	Trip 2	Responsibility
Ralph Baumgardner (EPA)	Denver-St. Louis	Los Angeles	Calibration
Gary Klauber (EMI)		Dallas-Raleigh	Operations
Keith Kronmiller (EMI)		Jackson-Raleigh	Operations, Engineering
Lee Langan (EMI)	Denver-St. Louis		Software, Operations
Gilbert Newcomb (EMI)	Denver-Kansas City		Engineering, Operations
Richard Paur (EPA)	Denver		Calibration
Michael Peache (EMI)	St. Louis-Raleigh		Operations, Meteorology
Philip Schug (EMI)	St. Louis-Lexington	Phoenix-Jackson	Engineering, Operations
Robert Stevens (EPA)	Kansas City-St. Louis		Calibration, Operations
William Vaughan (EMI)	Kansas City-Raleigh	Los Angeles-Phoenix	Operations, Calibrations
Charles White (EMI)		Los Angeles-Dallas	Software, Operations

Data reduction and report preparation were accomplished by Lee Langan, Michael Peache, Jean Jacques Garbarz, Joan Geary, and Evelyn More.

SECTION 1

INTRODUCTION

On two occasions in 1976, very sensitive, rapid-response air quality analyzers were driven across the United States to seek data background levels and to observe cross-urban effects. This report presents the results of these trips. The results may be of interest to those concerned with air quality in rural regions, with the fate of sulfur or nitrogen oxides, and with the establishment and monitoring of national standards.

The West-to-East traverses extended from Denver, Colorado, to Raleigh, North Carolina, in August, and from Los Angeles, California, to Raleigh in November. Nearly 8,000 kilometers were driven. The general routes are shown in Figure 1.

The surveys were conducted by Environmental Measurements, Inc. (EMI) with the support of two U. S. Environmental Protection Agency (EPA) contracts: Purchase Order DA-6-99-6876A and Contract 68-02-2484. The report was prepared under Purchase Order DA-7-3954A. The sensors included a total sulfur gas monitor, a dual-chamber nitrogen oxide and nitrogen dioxide monitor, and a state-of-the-art ammonia monitor.

Most of the data were gathered while moving, and an effort was made to stay away from nearby motor vehicles that could influence results. Of necessity each traverse extended over several days; the intent was to cover distances between major urban sites with continuous data. Generally, the day's operations began and ended with calibrations of all instrumentation. Periodically, on some days, stopped data were gathered at rural sites for a few hours.

The equipment and operating procedures are discussed first. A presentation of the results and the relevant meteorological conditions follows. The concluding sections (4 and 5) contain observations, discussion, and analyses.

This dual set of long-distance, *en route* measurements has demonstrated the value of moving environmental monitoring by this technique. The overview of the geographic distribution on a broad scale and the long distances of low levels interconnecting the urban anomalies depict the consistency of the air contamination. The regional plateaus of higher concentrations show the

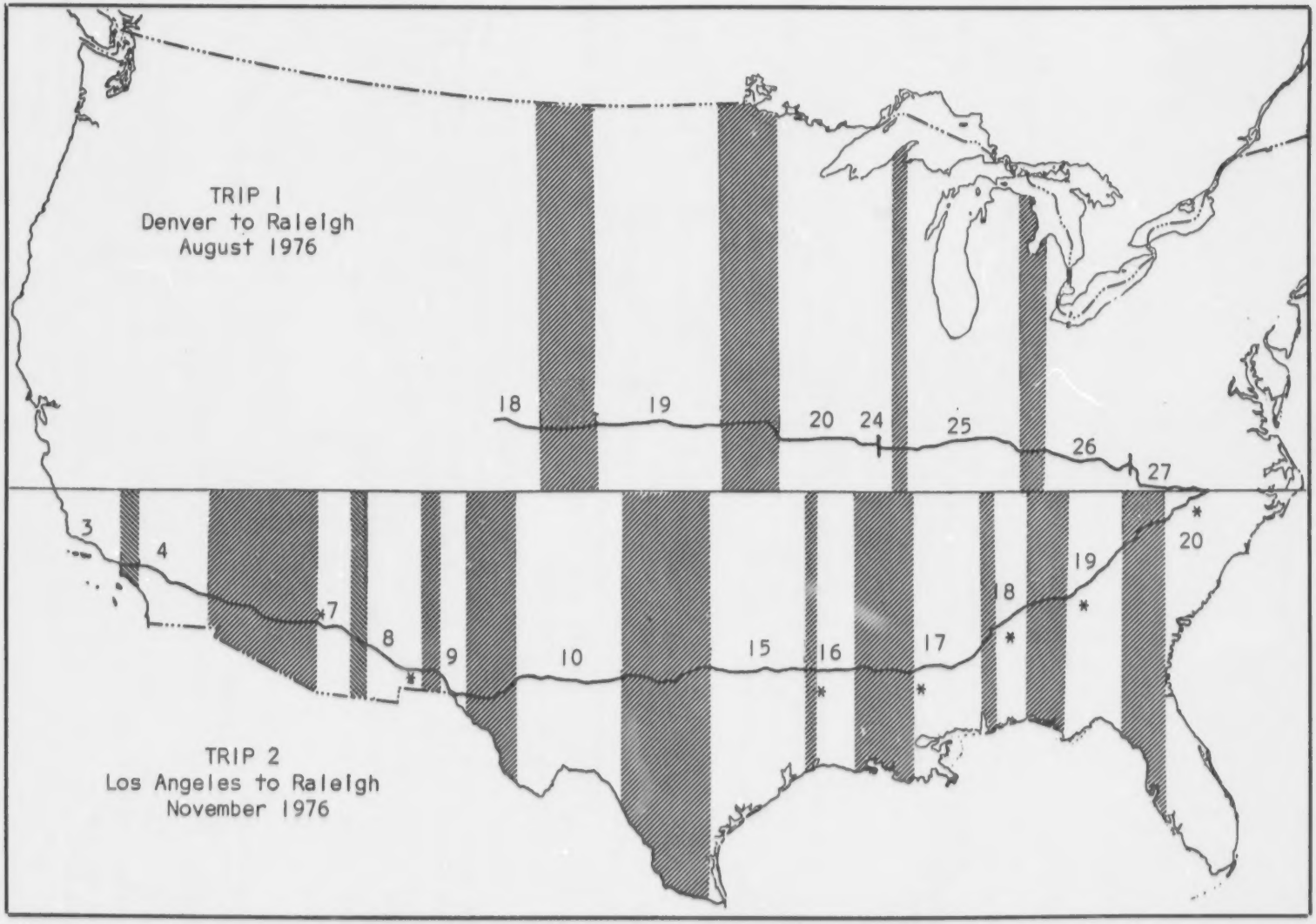


Figure 1. Routes of travel with each traverse day delineated. Shading indicates periods of monitoring while dark. Stopped data locations are noted by asterisk.

diffusion and collective gathering of many localized sources under low wind conditions. The background levels provide observations necessary for the use of regional modeling. The peaks and urban anomalies allow comparison of separate metropolitan sites using a rapid and relatively inexpensive reconnaisance method. Highly sophisticated instruments can be used in the field to cover large areas, to provide monitoring otherwise prohibitive in cost if a fixed-base network is established. And, if need be, anomalous conditions can be traced, at the time of their occurrence, by traversing up-wind of the initial contact.

SECTION 2

EQUIPMENT and OPERATING PROCEDURES

The EMI Air Quality Moving Laboratory (AQML) was an outfitted Dodge Maxivan designed for routine regional or local surveys. Data processing instrumentation was supplied by EMI; the high sensitivity analyzers and calibration and collection equipment were supplied by EPA.

The EMI/EPA staff developed operating procedures for the unique conditions of this continuous long-range measurement program. These ranged from the logistics of subsistence and maintaining a suitable pace to the details of quality assurance. Most operations became routine, making it apparent that such a technique of measurement could be maintained without undue stress, given a protocol and reliable equipment.

Data processing en route was automatic, requiring operator interaction only about each half hour. After the fact, editing and plotting were straightforward and included adjusting all traverses to a common coordinate system, reducing all measurements to common stored engineering units, and plotting the results.

The large data base suggested the use of larger computer memories. Fortunately, these have evolved with desktop computers since the project's field work. A Hewlett-Packard Model 9845B was used for final data analysis.

AIR QUALITY MOVING LABORATORY

The Dodge Maxivan used for the cross-country measurements is pictured at one of the stationary stop sites in Figure 2. This vehicle contained a standard support installation: an inboard 3.5kW Onan generator, an EMI MAP-II navigational and data system, two automotive air conditioners, desk space with four below-table racks for analyzers, a glass and Teflon air intake manifold with pump, a storage closet for operating and calibrating gases, and a Hewlett-Packard 9830A programmable calculator with peripheral printer, plotter, and tape cassette.

To this array were added EPA's analyzers: a Meloy Model 285 flame photometer total sulfur monitor, a Thermo Electron chemiluminescent NO/NO_x monitor (trip 1) and Aerochem chemiluminescent

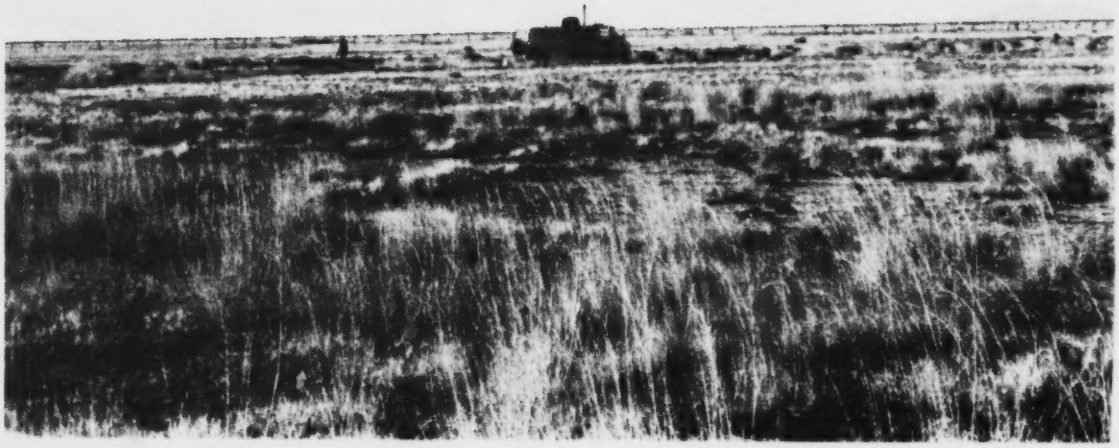
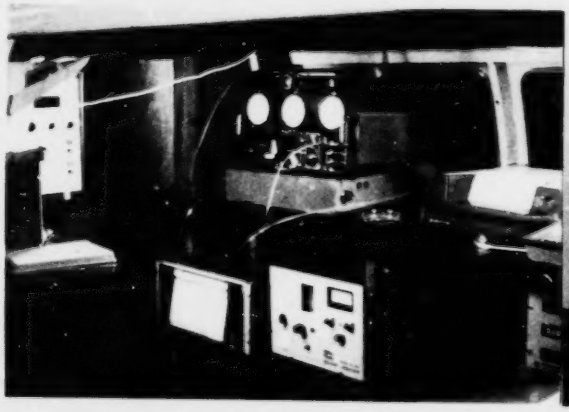


Figure 2. The EMI AQML proceeded a short distance off a New Mexico highway to gather stopped data near Lordsburg (8 November 76).

NO/NO_x ammonia monitor (trip 2), and related calibration equipment and gases. Some chart recorders and test instruments were also present. Figure 3 shows some of the equipment inside the van. The AQML proved sufficiently comfortable with two air conditioners, but it was crowded when four people were present, along with their luggage for the long trip.

The EMI Air Quality Moving Laboratory

HP9830, MAP-II System, and plotter inside the moving van



Meloy analyzer (below) and calibrating apparatus next to the HP9830 printer

Cross-calibration at EPA-RAMS Site 108

Figure 3. The Air Quality Moving Laboratory and interior details.

THE MAP SYSTEM

The EMI MAP II navigational and data system operates under the control of the HP-9830, which provides all data formating and printing. Each of the analyzers was connected to an analog-to-digital channel of MAP; sampling was performed each 20 meters along the route of travel. Each 200 meters the average measurement was listed, together with the position and time of the last sample, and plotted in realtime. While the AQML was stopped, three-second samples stored every 30 seconds were obtained. These data were stored on tape cassettes for subsequent analysis; they were also plotted in realtime for review by the operator. When only two analyzers were in use, switching on the chemiluminescent monitor and scrubbing on the flame photometric monitor were noted by automatic formating changes in the data listings.

A partial list from trip 1 (shown in Figure 4) is typical of the visual data. Each *line* contains location, time, digitized analyzer data, and the difference in seconds and meters since the preceding line. Each *event* covered a grouping of consistent data (between two known points, stopped data, a calibration, and the like). Because of the limit of the 9830's memory, no event of 200-meter data exceeded 50 kilometers (250 lines of data). En route, this was about an hour of data, referred to as a *set* of data. Each cassette could store six sets of data, so more than two cassettes were rarely used for each day's data.

In addition to the data averaging, the MAP computer derives the actual position of the vehicle each meter along the route. By combining this position with the bearing of the vehicle, monitored by the gyrocompass installed in the van, a new Cartesian (x,y) coordinate is calculated. These north-south and east-west distances are added to a position defined at the beginning of each event. Every 200 meters a new position is identified.

A quasi-Universal Transverse Mercator (UTM) system of geographic locations was attempted because values from this accepted framework were used at the beginning of each trip. The UTM reference is not, however, contiguous across the country. Due to the earth's curvature, several grids intersect. The initial UTM reference was simply continued for each trip so that the plotted routes that follow are equal-area projections across the United States.

MEASUREMENT INSTRUMENTATION

A high resolution total sulfur monitor manufactured by Meloy Laboratories, Inc. was installed in the AQML. Sensitivities on the order of one part per billion (ppb) were obtained. A chemiluminescent monitor manufactured by the Thermo Electron Corporation was installed to measure total nitrogen oxides (NO_x)

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EVENT # 26 DAY 760825 CODE: 23040 WIND: 0.0 M/S FROM 45 DEG

0 0 =SOURCE ---------BACKGROUND----------
613000 4240000 =ORIGIN 13 0 0 0 0 0

	X	Y	TIME	#1 TS	#2 TECO	#3 NO	#4 NOX	#5 NH4	#6	D	T
48	611936	4241245	195003	48	124	124	0	0			
49	612270	4241079	195813	49	147	147	0	0		380	490
50	612446	4240935	195828	53	172	172	0	0		200	15
51	612565	4240830	195841	54	212	212	0	0		200	13
52	612570	4240631	195856	53	232	232	0	0		200	15
53	612581	4240432	195909	52	209	209	0	0		200	13
54	612679	4240259	195923	51	195	195	0	0		200	14
55	612780	4240088	195947	41	194	194	0	0		200	24
56	612917	4239947	200011	51	185	185	0	0		200	24
57	612962	4239865	200035	58	195	195	0	0		200	24
58	612957	4239927	200101	57	212	212	0	0		200	26
59	613144	4239992	200114	59	204	204	0	0		200	13
60	613311	4240100	200126	58	200	200	0	0		200	12
61	613475	4240212	200138	59	205	205	0	0		200	12
62	613640	4240321	200149	60	212	212	0	0		200	11
63	613806	4240433	200200	61	224	224	0	0		200	11
64	613958	4240559	200211	60	227	227	0	0		200	11
65	614105	4240694	200222	61	229	229	0	0		200	11
66	614252	4240827	200235	61	229	229	0	0		200	13
67	614437	4240887	200249	60	237	237	0	0		200	14
68	614609	4240797	200307	59	240	240	0	0		200	18
69	614714	4240630	200328	57	230	230	0	0		200	21
70	614815	4240457	200348	57	217	217	0	0		200	20
71	614916	4240287	200408	57	205	205	0	0		200	20
72	614943	4240285	200634	58	189	189	0	0		200	140
73	614848	4240460	200711	58	165	165	0	0		200	37
74	614751	4240634	200814	56	159	159	0	0		200	6
75	614658	4240809	200830	59	150	150	0	0		200	1
76	614630	4241002	200844	58	159	159	0	0		200	14
77	614742	4241161	200856	56	177	177	0	0		200	12
78	614907	4241270	200907	55	174	174	0	0		200	11
79	615073	4241381	200918	56	162	162	0	0		200	11
80	615239	4241490	200928	58	165	165	0	0		200	10
81	615406	4241598	200938	59	177	177	0	0		200	10
82	615575	4241701	200948	59	190	190	0	0		200	10
83	615748	4241800	200959	58	200	200	0	0		200	11
84	615920	4241898	201009	57	199	199	0	0		200	10
85	616102	4241981	201022	55	197	197	0	0		200	13
86	616279	4242073	201036	54	204	204	0	0		200	14
87	616470	4242047	201051	55	204	204	0	0		200	15
88	616668	4242070	201105	55	204	204	0	0		200	14
89	616866	4242044	201118	53	209	209	0	0		200	13
90	617065	4242022	201132	52	210	210	0	0		200	14
91	618026	4241989	201242	43	197	197	0	0		965	70
92	617964	4242102	201330	46	217	217	0	0		235	48
93	617862	4242273	201345	45	244	244	0	0		200	17
94	617761	4242443	201400	42	237	237	0	0		200	15
95	617669	4242619	201413	40	222	222	0	0		200	13
96	617609	4242810	201425	38	219	219	0	0		200	12
97	617560	4243004	201439	36	217	217	0	0		199	14
98	617497	4243192	201452	36	214	214	0	0		201	13
99	617400	4243365	201506	37	209	209	0	0		200	14
100	617297	4243536	201525	37	217	217	0	0		200	19
101	617296	4243597	201607	32	217	217	0	0		155	42
102	617296	4243597	201615	30	217	217	0	0		0	8

TIME: 20.2 MN DISTANCE: 11.5 KM

TS CAL= 0.153 TECO CAL= 1.470

Figure 4. HP-9830 printout of MAP system averages in the distance mode (every 200 meters) showing the location (x,y), the time, the total sulfur (TS), the values for NO or NO_x from the TECO readings, the distance (D) between two averages, and the time (T) between those averages.

and nitric oxide (NO) in August; a prototype chemiluminescent monitor produced by Aerochem Research Laboratories was used in November. Bendix Corporation and Research Triangle Institute double-dilution calibration systems were used for spanning the low level concentrations of both systems; a sulfur dioxide permeation tube and NO gas cylinder were concentration sources. Ultra high purified zero air produced by Scott Merrin, Inc. was used to attain zero levels to monitor potential instrumental drifts at these low levels. Frequent "zeroes" in the total sulfur were also obtained by using a Meloy SO_2 scrubber manufactured by Meloy.

Background gaseous sulfur measurements were made with the Meloy Model SA-285, which uses the flame photometric detector as its measurement principle. The measurement is of the chemiluminescense produced when gaseous sulfur compounds are burned in a hydrogen-rich flame. The reaction emits a broad band of light with its maximum at 394nm. The intensity of the emission is directly proportional to the square of the sulfur concentration.

This monitor is designed to have optimum sensitivity and stability to allow for measurements as low as 1 ppb. Sample air is drawn continuously through the detector at a controlled flow rate of 200cc per minute. Hydrogen is applied to the detector at a positive flow of 150cm^3 per minute. The light emission is measured with a cooled photomultiplier tube. The current output of the photomultiplier tube is linearized and connected to a voltage before display on a meter or recorder. By using selective chemical scrubbers supplied with the unit, it is possible to remove hydrogen sulfide (H_2S) or sulfur dioxide (SO_2). By measuring the total sulfur present, then placing the appropriate scrubber in line, it is possible, by measuring the difference in signal, to determine specifically the SO_2 or H_2S.

Measurements of background levels of NO and nitrogen dioxide (NO_2) were made during the August trip with a Thermo Electron Model 12 NO-NO_x monitor. This instrument can measure NO and NO_2 from 0 to 1000 ppb with a detectable minimum of 2 ppb by using the gas phase chemiluminescent reaction of nitric oxide and ozone. A high temperature thermal converter converts NO_2 to NO, allowing for measurement of NO_x (NO_2 + NO) and the determination of NO_2 present by subtraction (NO_x - NO).

Sample air is pulled through the NO detector at a flow rate of 100cc per minute where it reacts with ozone at a reduced pressure of 10 torr. A switching valve gives a response for NO; then the sample is sent through the thermal converter for a NO_x (NO + NO_2) measurement. A red-sensitive photomultiplier tube with an optical filter observes the light from the reaction of NO to O_3 in the detector and converts it to an electrically amplified signal. An output signal for both NO and NO_2 is possible.

Measurement of background levels of NO and NO_2 were made during the November trip with a prototype nitrogen dioxide, nitric oxide, and ammonia monitor manufactured by Aerochem Research Laboratories. This monitor also uses the reaction of NO and O_3 as its measurement principle. Using a detector especially designed by Aerochem to increase the residence time for the NO:O_3 reaction, the Aerochem monitor is capable of measuring NO, NO_2, and ammonia (NH_3) from 0 to 500 ppb with minimum detectable limit of 1 ppb. The monitor has two NO:O_3 detectors, each capable of measuring NO or NO_x by using a thermal converter. The monitor was set up for the study with one channel having a low temperature (300°C) converter in line to measure NO + NO_2 and the other channel having a high temperature (1000°C) converter to convert NO_2 and NH_3, giving a signal for NO, NO_2, and NH_3. The sample air is pulled into the monitor at a flow of 500cc per minute, half of the flow going to each detector. Ozone flow for the reaction is generated, using a single ozone generator with the flow being split into each detector.

Using thermal converters with each detector and an ammonia scrubber with one detector, it was possible to measure background levels of NH_3 by subtracting the difference between the signal with the scrubber and the signal generated without the scrubber. These results were monitored only experimentally during the trips.

CALIBRATION

At the beginning of the project, an effort was made to calibrate the measuring instruments each morning and evening. This allows for conversion of response units to convenient and familiar units: parts per billion (ppb). As the program continued, at least one calibration was made each day (Figure 5).

Two calibration systems were used during the studies. The first, a Bendix Model 8851 calibration system, has capabilities for generating known concentrations of NO and NO_2 (using the gas phase titration of NO with O_3) and of sulfur compounds and NH_3 (using permeation tubes). A cylinder of NO at 49 ppm (standardized by the National Bureau of Standards' Standard Reference Method) was diluted to produce concentrations from 0.01 ppm to 0.50 ppm using the Bendix system, which has dilution flow control up to 8000cc per minute. The chemiluminescent NO-NO_x monitors were calibrated for NO by this technique.

The Bendix system also has a permeation tube oven that provides controlled temperature to within 0.1° at 25°C. Dilution air passes through the oven housing the permeation tubes of either SO_2, H_2S, or NH_3. Permeation rates for each of these tubes were determined gravimetrically before the beginning of the study.

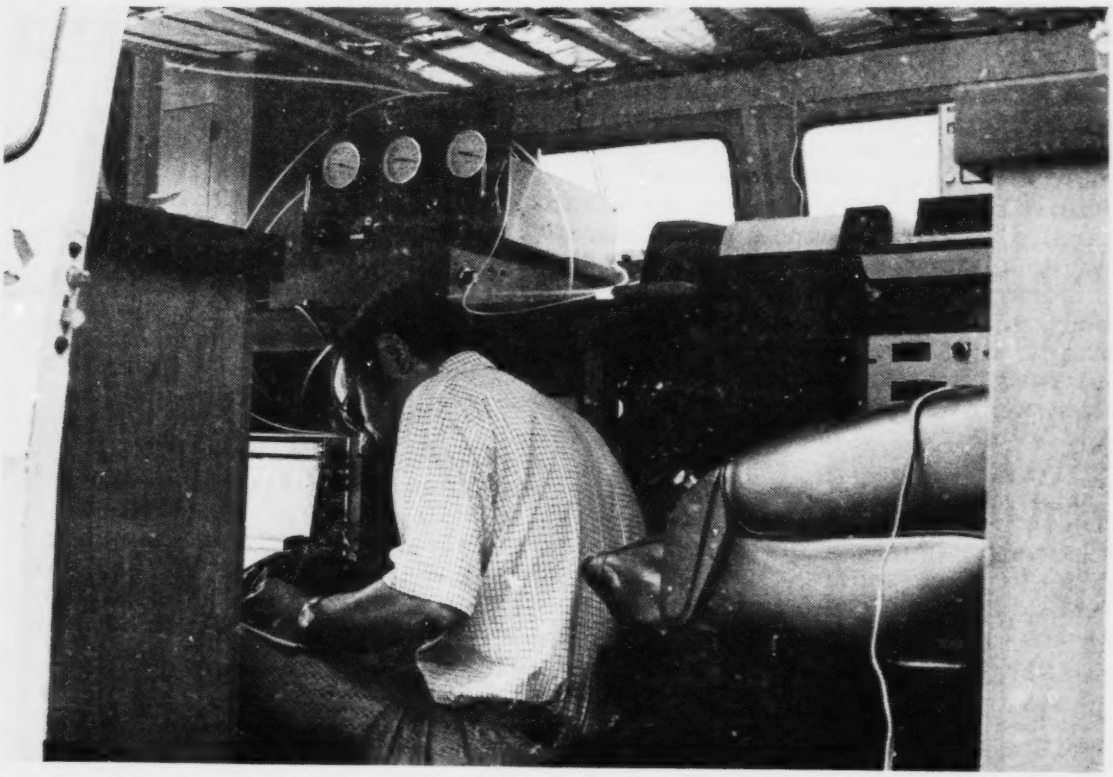

Figure 5. Calibrations were normally accomplished twice a day, before and following the travel.

Using the permeation system of the Bendix, calibration concentrations of SO_2, H_2S, and NH_3 between 0.010 and 0.50 ppm could be generated.

The second calibration system used in the study was a double dilution permeation system built by Research Triangle Institute to deliver known concentrations of gases below 0.010 ppm. Using the RTI system, known concentrations of SO_2, H_2S, and NH_3 were generated between 0.001 and 0.010 ppm. The RTI permeation calibration system consists of two automatic flow controllers, a temperature-controlled permeation tube oven, and an automatic digital readout of flows and temperature. Two mass-flow controllers control air flow of the permeation oven and dilution flow between 200 and 10,000cc per minute. Zero air entering the unit is split, part going through one flow controller to the permeation oven, providing flow over the permeation tube, and part going through the second flow controller, providing dilution flow. A glass capillary allows 100cc per minute of air flowing over the permeation tube to be mixed with the dilution air, giving a double dilution to the original concentration. Temperature within the permeation oven is controlled to within 0.10°C. The double dilution system was used

to achieve very low concentrations for SO_2 for calibration across the usual ambient range observed. Careful selection and calibration of the flows with a bubble tube resulted in three-point calibrations ranging from 10 to 25 ppb of SO_2. The instrument received at least one three-point calibration every survey day and one or two span checks at the 25-ppb level to confirm the stability of instrumental calibration under the vibration and temperature conditions experienced in the AQML. Although there was some zero drift observed with occasional high temperatures, it amounted to fewer than 7 to 8 ppb SO_2 and was confirmed by monitoring zero air, as opposed to mere SO_x scrubbing. These drifts have been removed from the data. The stability of the calibrations was excellent during the course of the study, requiring no changes in calibration factors over a given week of moving measurements.

Periodically an SO_x scrubber was inserted into the sample line to confirm that the principal signal was SO_2. Generally, SO_x scrubbing brought the instrument response to within 2-3 ppb of zero air readings, indicating that the majority of sulfur was coming from SO_x and not H_2S. The only place an appreciable (10 ppb) H_2S signal was observed with the SO_x filter in place was in Eastern Texas and Western Louisiana.

OPERATIONS

After the morning calibration the AQML was driven along the least-travelled route to the next major urban area. Individuals shared the load of driving and monitoring the data gathering. Periodic interaction was required with the automated system, route planning, and usual observations. The intent was to traverse interurban regions in a continuous survey. A conscious attempt was made to stay away from traffic or, when this proved difficult, to stay away from any truck or automobile by either passing or dropping back. The measured effect of an adjacent vehicle's pollution is obvious in the data plots. Data spikes are common but of short duration. They have not been removed from the data. Diesel trucks created larger anamolies in nitrogen and sulfur oxides than did gasoline-powered automobiles.

Seven times during the second trip the AQML was stopped to make stationary measurements in conjunction with filter samples taken at the same time and analyzed for NH_3 by the Air Monitoring Center of Rockwell International, as described in Appendix A. The sites chosen to make the stationary measurements were several kilometers off the highway in rural areas. Stationary and filter data were collected near Lordsburg, New Mexico; Shreveport, Louisiana; Jackson, Mississippi; Ragland, Alabama; Commerce, Georgia; and Silver City, North Carolina. In addition, one set of stationary data only was collected near Superior, Arizona.

Included in Appendix A is a letter from Willard Richards of Rockwell International describing the filter analysis, along with a table of the results.

Summary maps of meteorological conditions are plotted in Figures 6 and 7. The conditions were noted during the traverses, but the overview was derived from the U.S. Weather Bureau's daily maps representing the 0700 EST status. The data plotted are taken from these publications and display the condition most representative and nearest the moving laboratory as it proceeded across the country. The 0700 wet and dry bulb temperatures and the 24-hour variation for the day are plotted in Figure 6 for each of the 18 survey days; upper-level wind speeds and directions complement these. In Figure 7 ground winds and the movement of regional highs are plotted to reflect forces on the pollution movements.

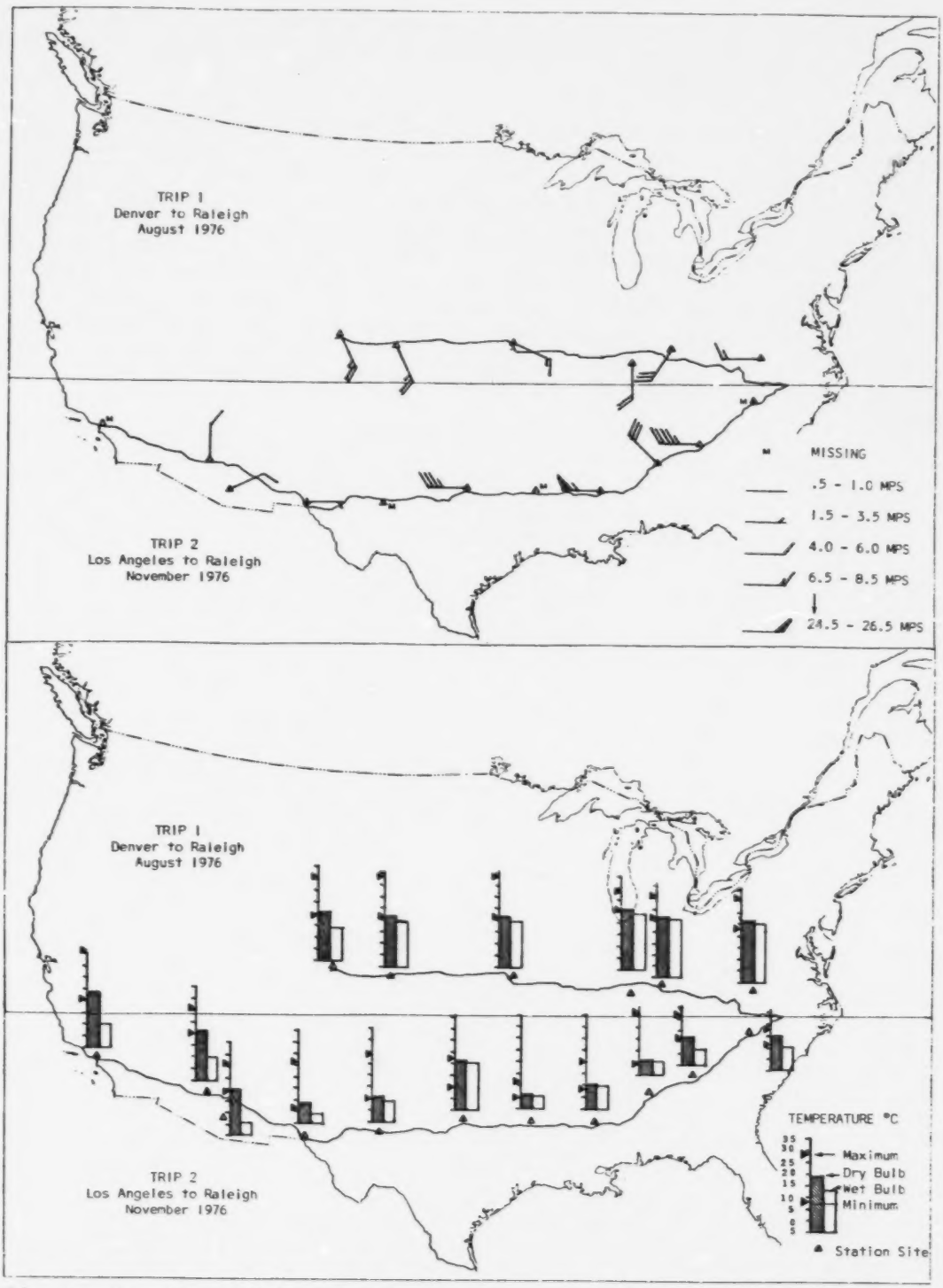

Figure 6. Ground-level temperatures and 500 mb wind direction and speed plotted for 0700 EST each day.

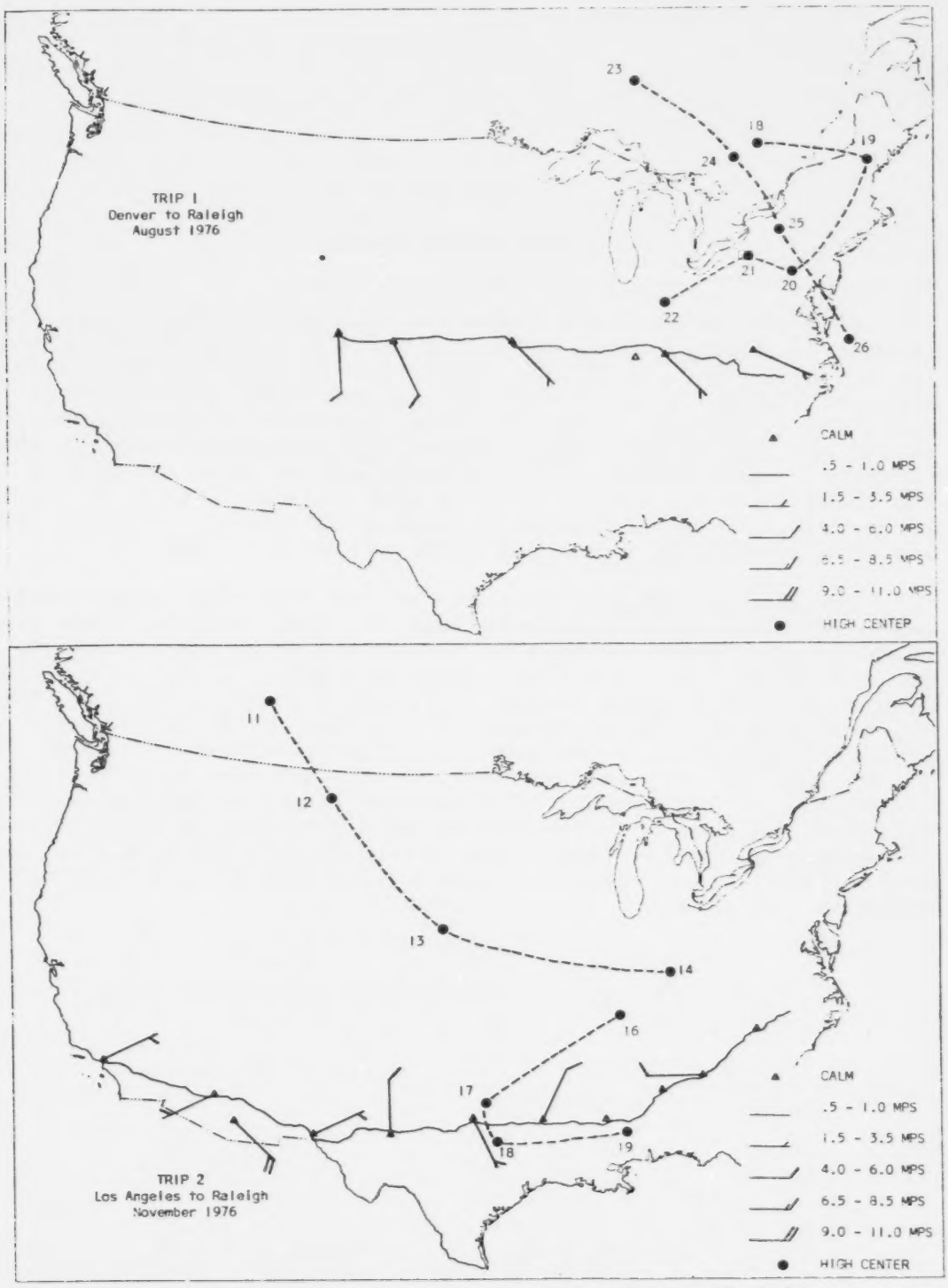

Figure 7. Surface wind direction and speed and daily movement of major high pressure centers influencing measurements.

SECTION 3

DATA PRESENTATION

Most of the data are presented in a two-figure format, one per sampling day. Total sulfur is on each top graph and nitrogen oxides on the one below. On both, the road has been plotted above the data. Locations of cities and times are indicated along the roads. On those occasions when the distance covered during the day of sampling exceeded the maximum allowed by the plotting frame (440km), the remainder of the data was placed in the right hand top corner of each graph. The figures, 8 through 26, conclude this section in chronological order. Figures 27 and 28 are the trip summaries from preliminary reports.

Scrubbing of SO_x is apparent on the data plot when sharp vertical lines fall to 3ppb or less of total sulfur. Data during stopped measurement periods of the second trip are listed in Table 2 below. In each case, these are rural sites several kilometers off the highway (refer to Figure 2 on page 5), and they are composed of sets of 30-second averages (3-second samples) over an approximately two-hour mid-afternoon period. About 30% of the time the SO_2 was scrubbed, which provided a total sulfur result and an SO_2 result. The stopped data are spatially indicated on the road and are plotted as a function of time in Figures 29 through 35, which follow the moving data. The total sulfur is above the nitrogen oxides; scrubbing and NO_x-NO switching are shown in the data.

TABLE 2. STOPPED DATA SUMMARY

DATE	SITE (near)	TOTAL SULFUR (ppb)	SO_2 (TS-scrubber) (ppb)	MEASUREMENT PERIOD (minutes)
7 Nov 76	Superior, AZ	1.78	1.27	63.8
8 Nov 76	Lordsburg, NM	1.16	1.16	180.6
15 Nov 76	Shreveport, LA	7.23	3.57	122.3
17 Nov 76	Pelahatchie, MS	12.47	6.92	127.2
18 Nov 76	Ragland, AL	11.25	5.30	120.6
19 Nov 76	Commerce, GA	9.41	1.76	126.9
20 Nov 76	Silver City, NC	22.24	15.89	161.1

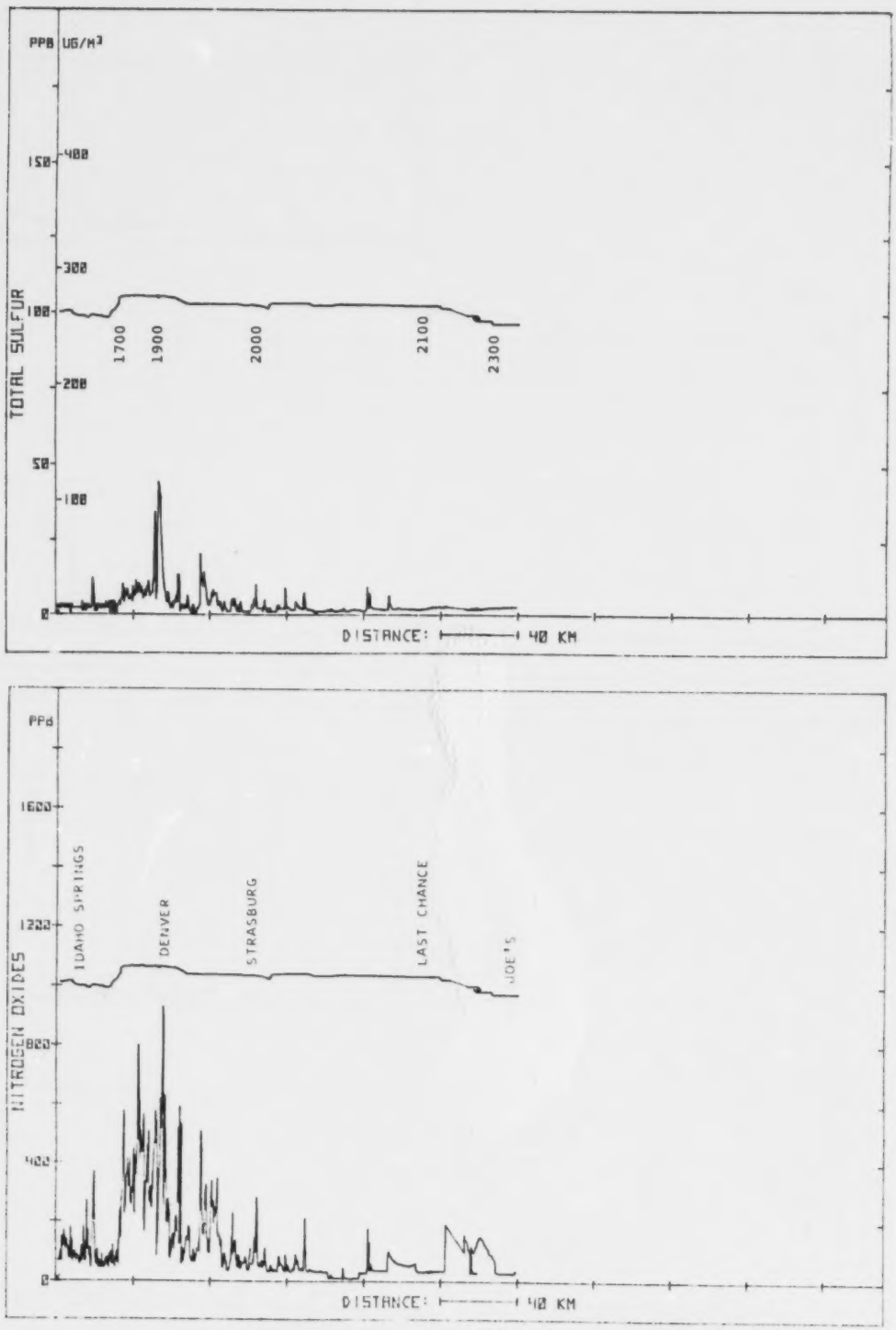

Figure 8. August 18 - 1600-2300 MDT - Colorado

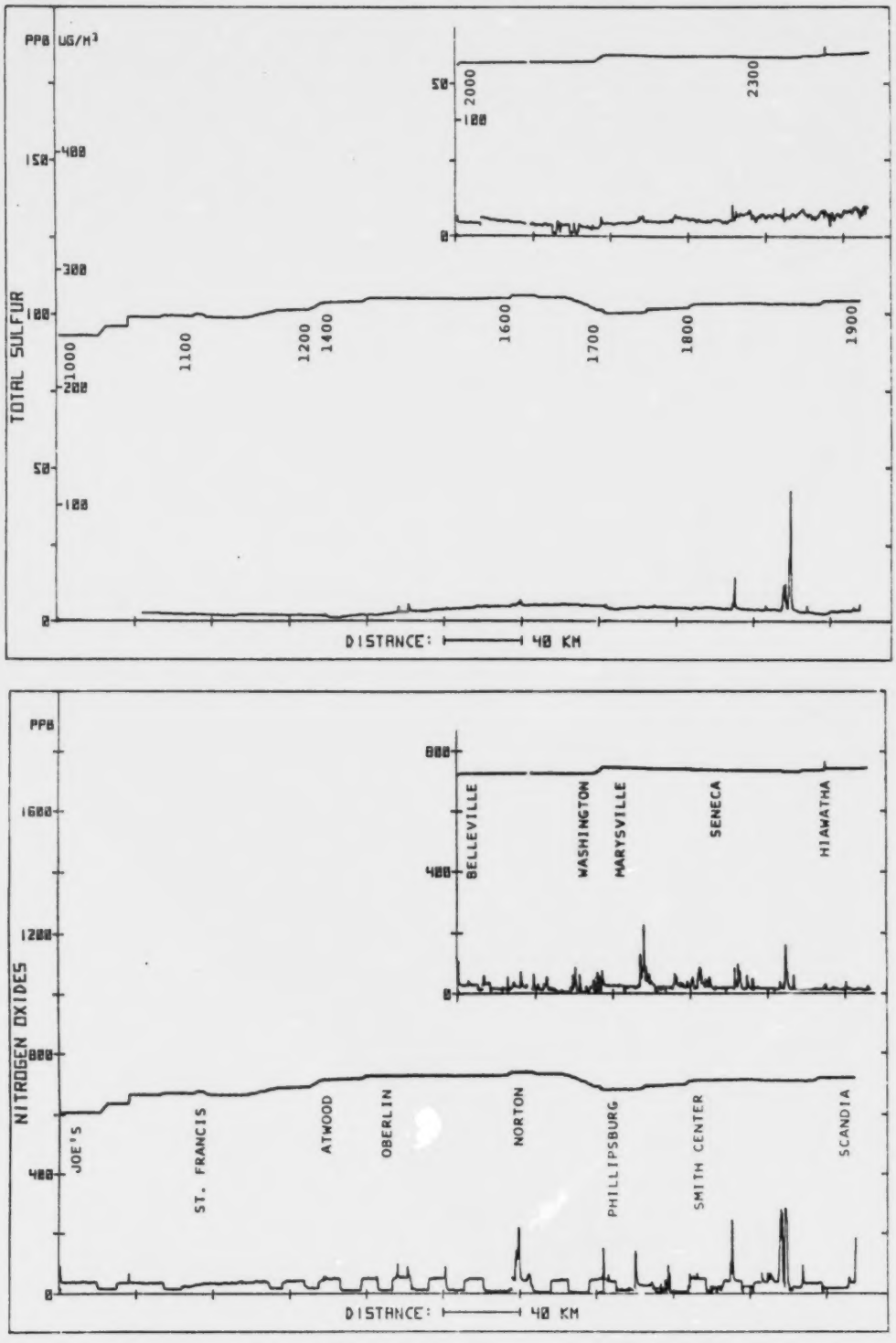

Figure 9. August 19 - 1000- 2400 MDT - Colorado-Kansas

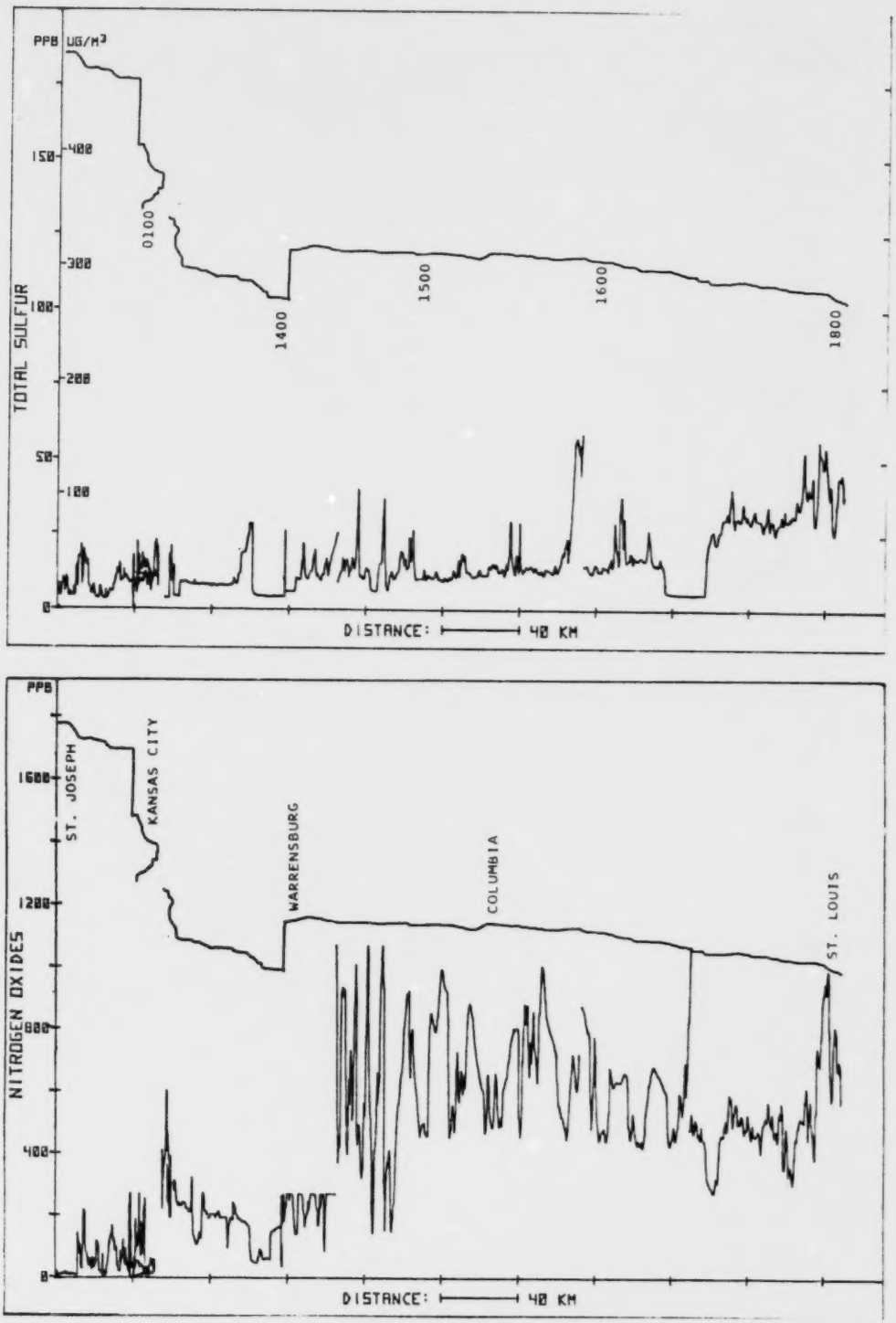

Figure 10. August 20 - 0000-0100, 1300-1800 CDT - Kansas-Missouri

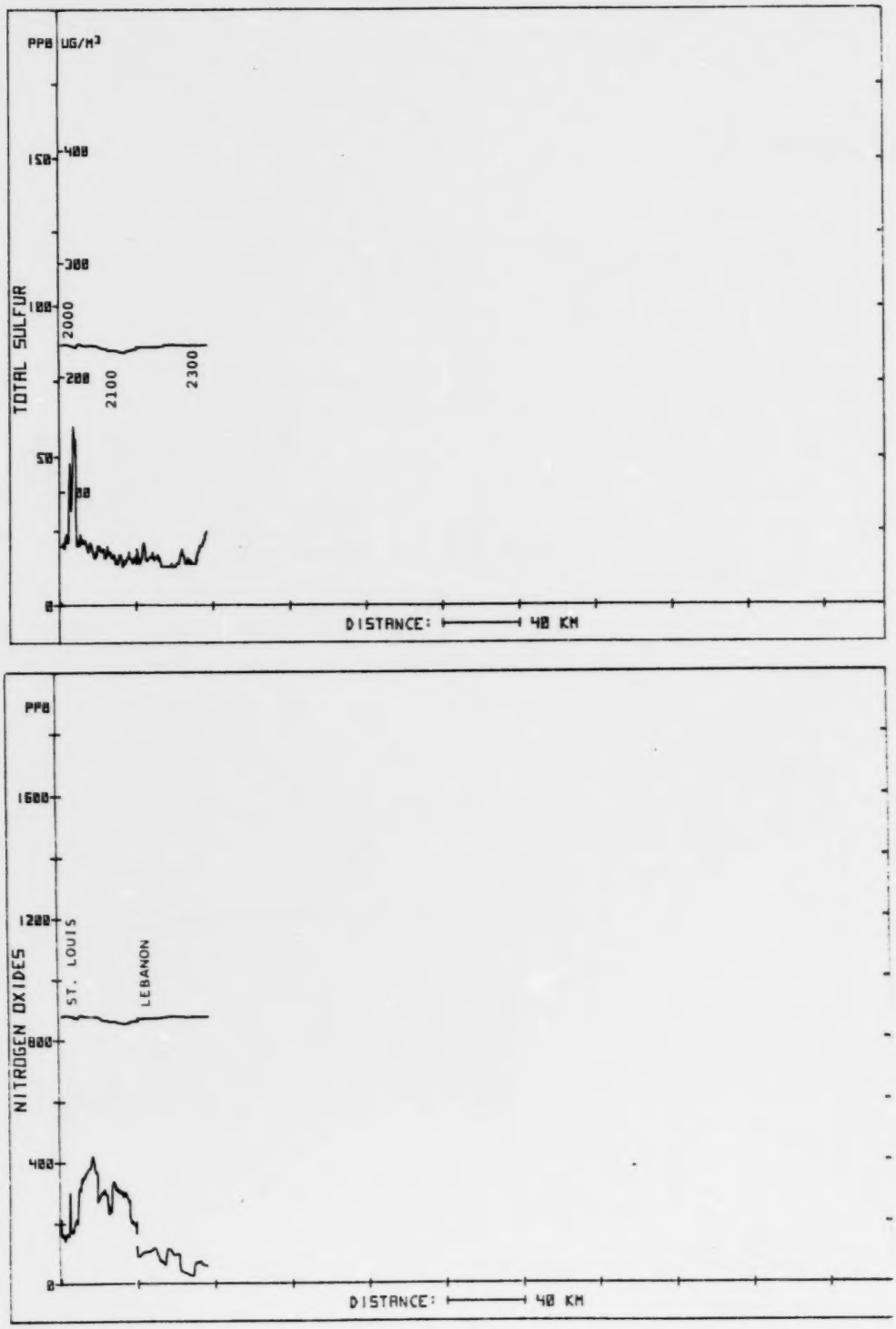

Figure 11. August 24 - 2000-2300 CDT - Missouri-Illinois

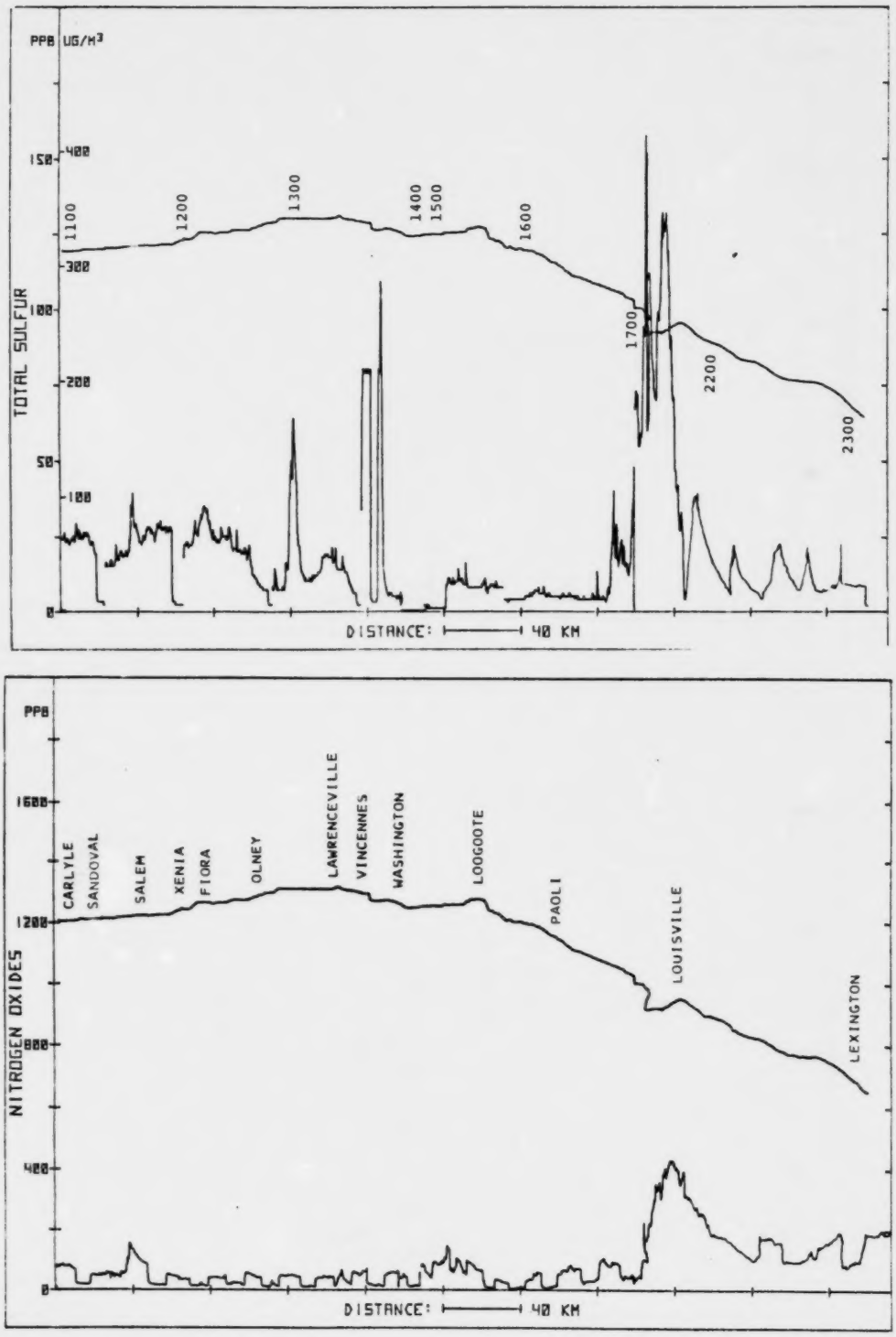

Figure 12. August 25 - 1100- 2300 CDT - Illinois-Indiana-Kentucky

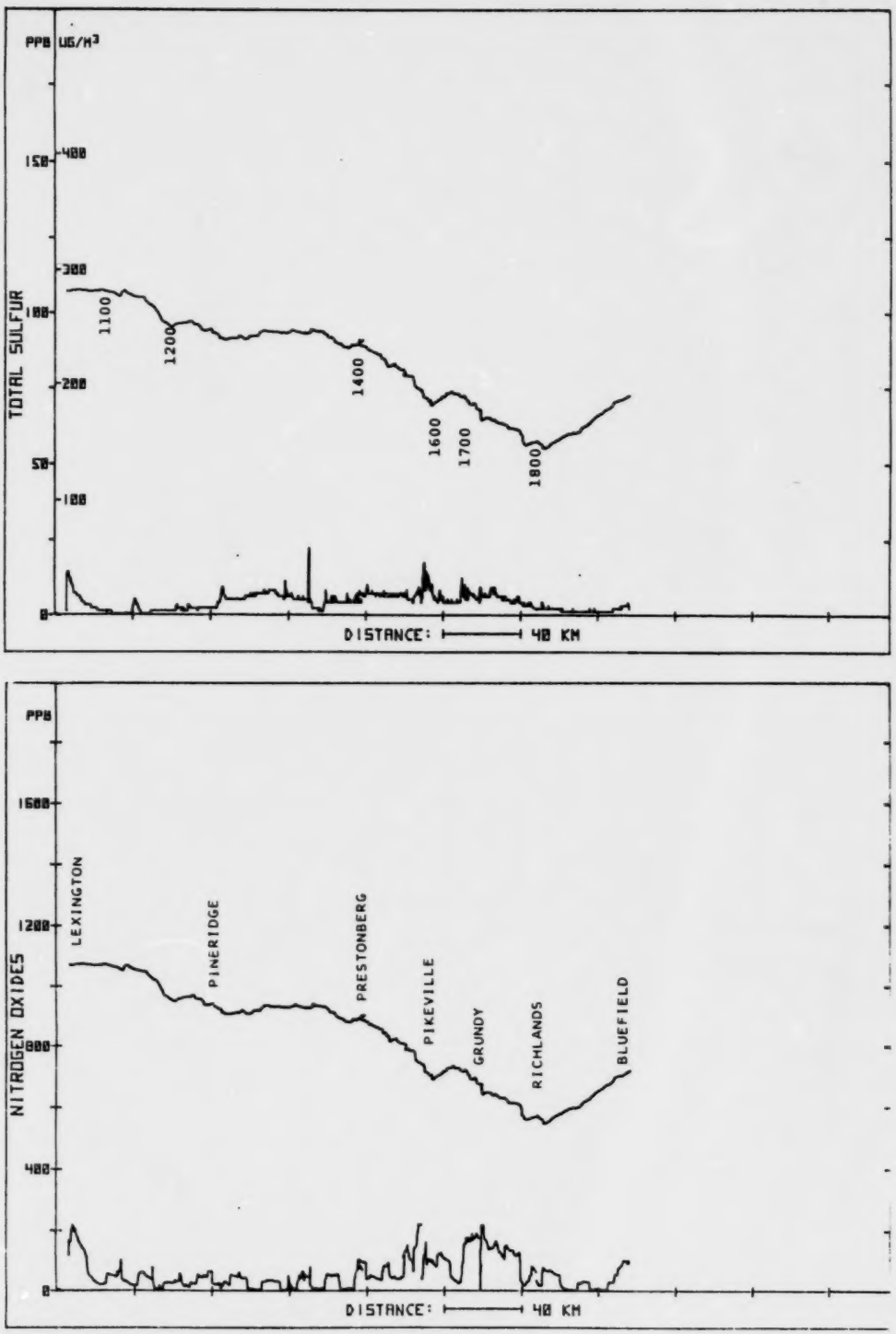

Figure 13. August 26 - 1000-1900 EDT - Kentucky-Virginia

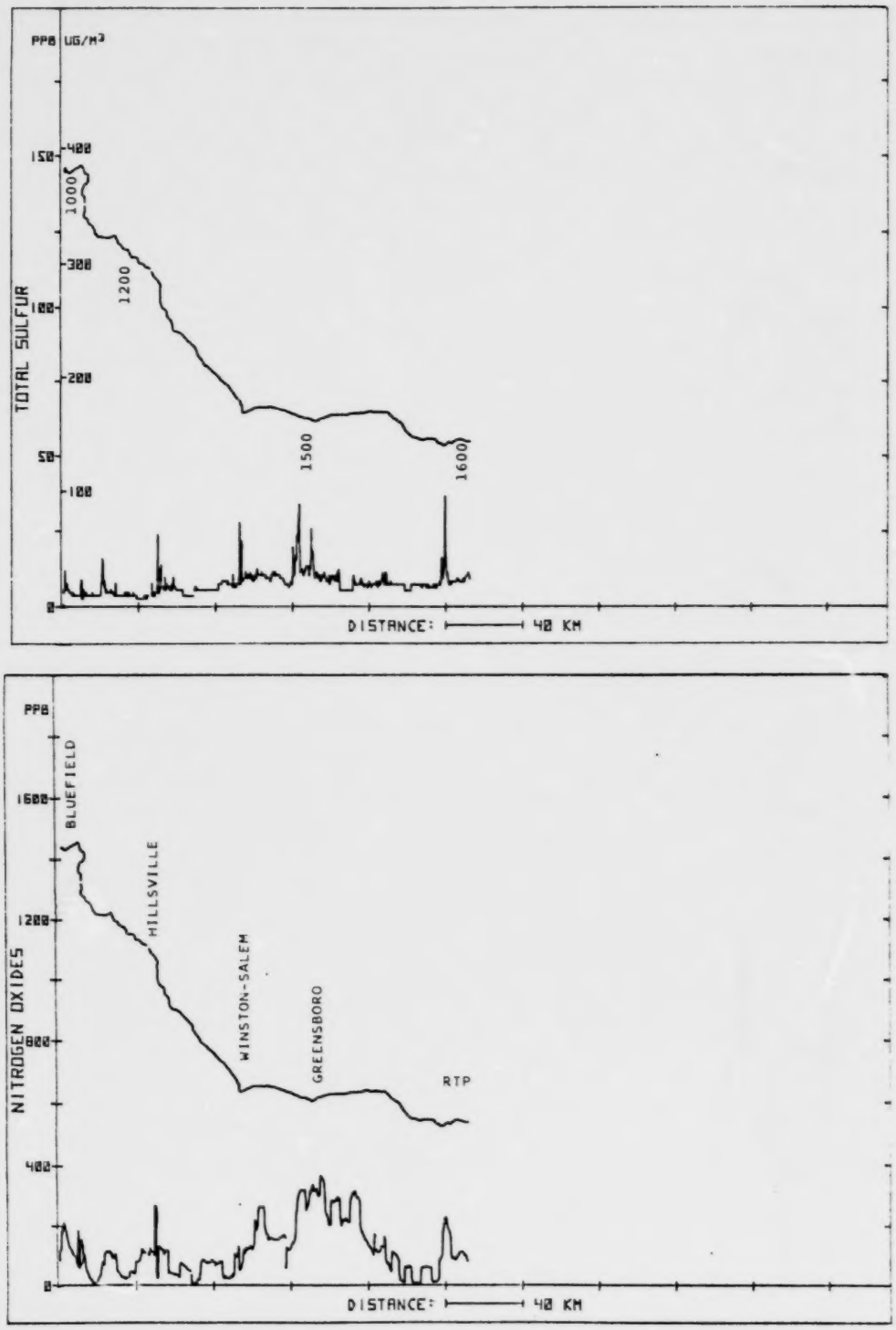

Figure 14. August 27 - 1000-1600 EDT - Virginia-North Carolina

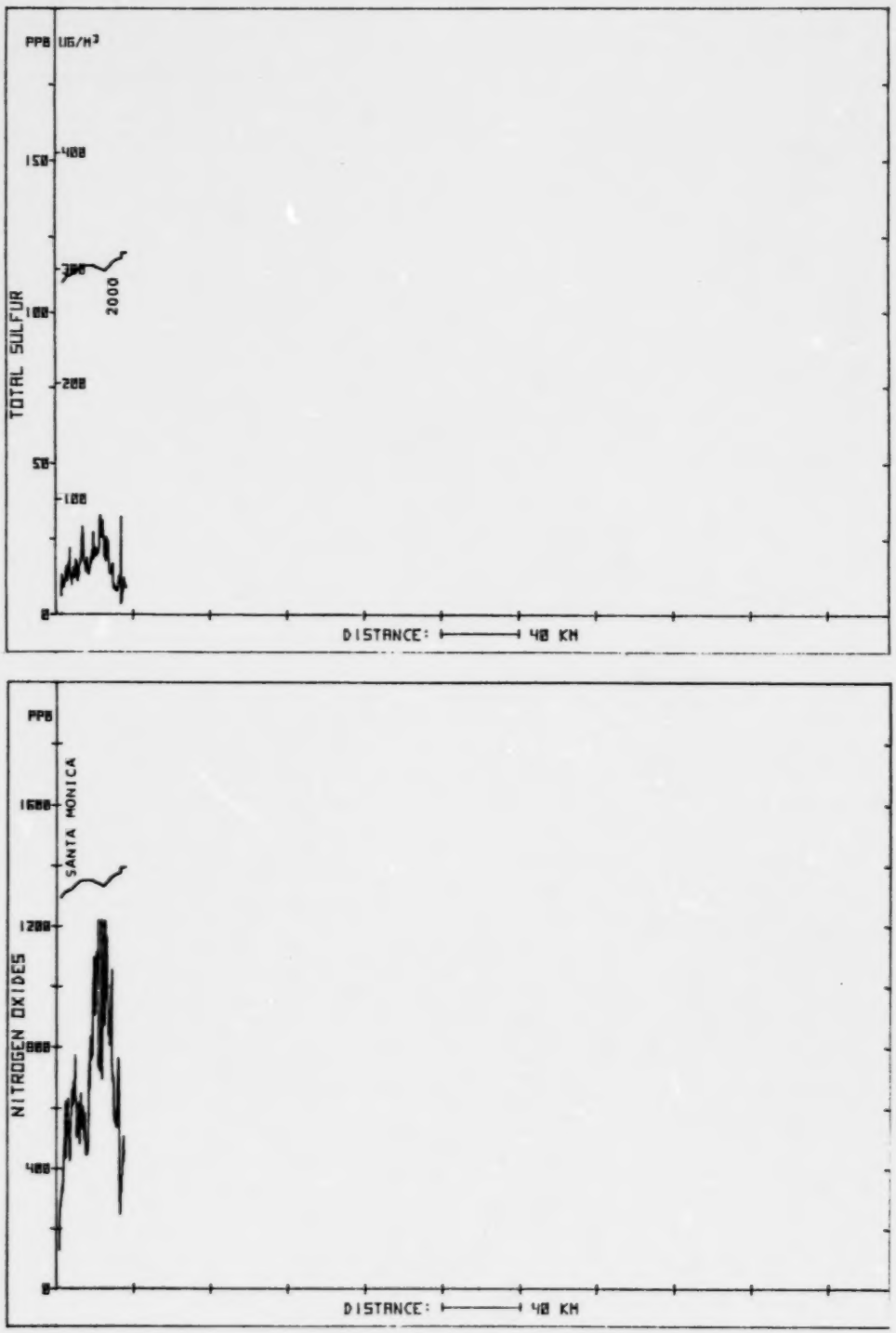

Figure 15. November 3 - 1900-2100 PST - Los Angeles

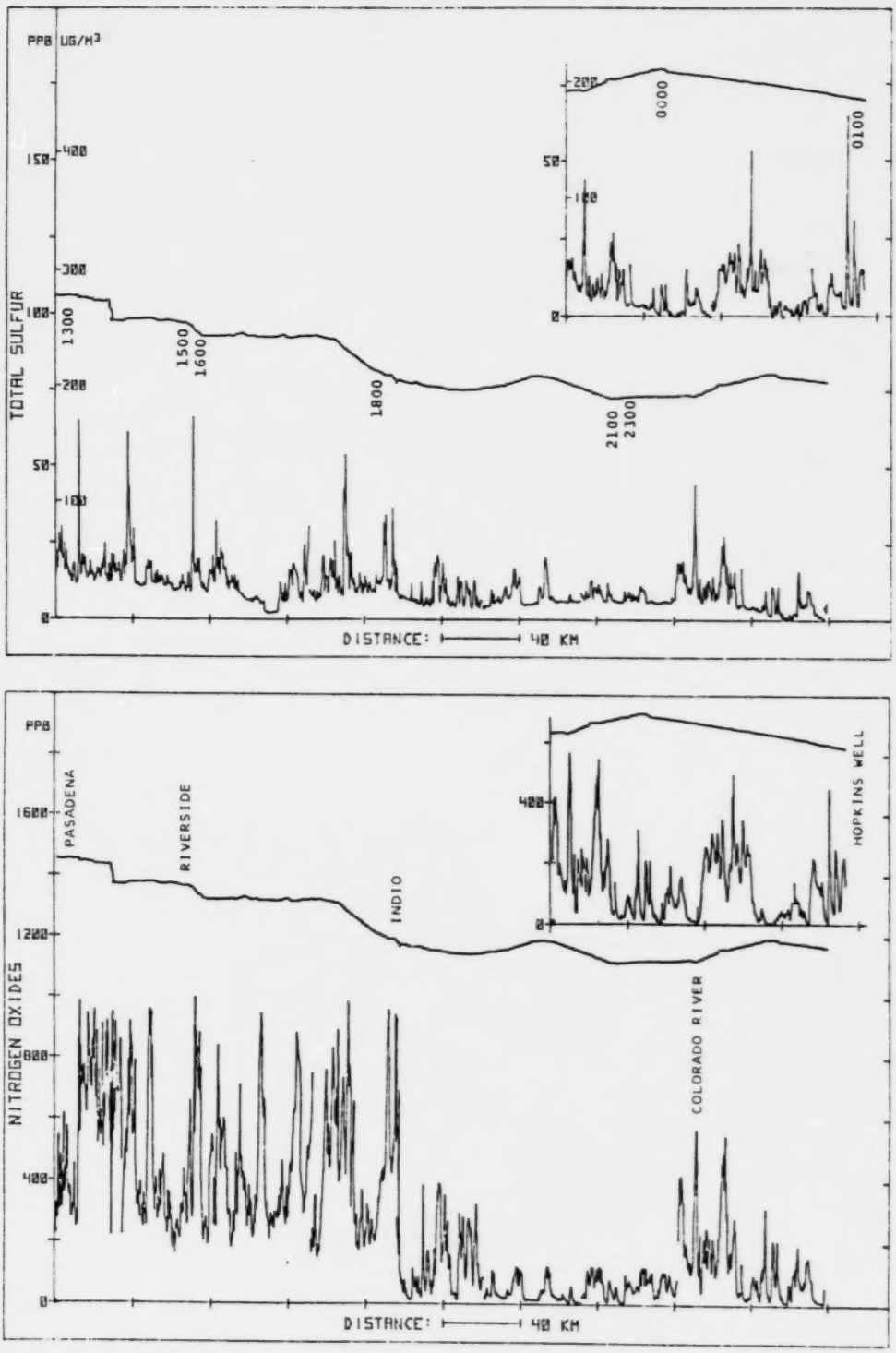

Figure 16. November 4 - 1300 PST to 0200 MST - California-Arizona

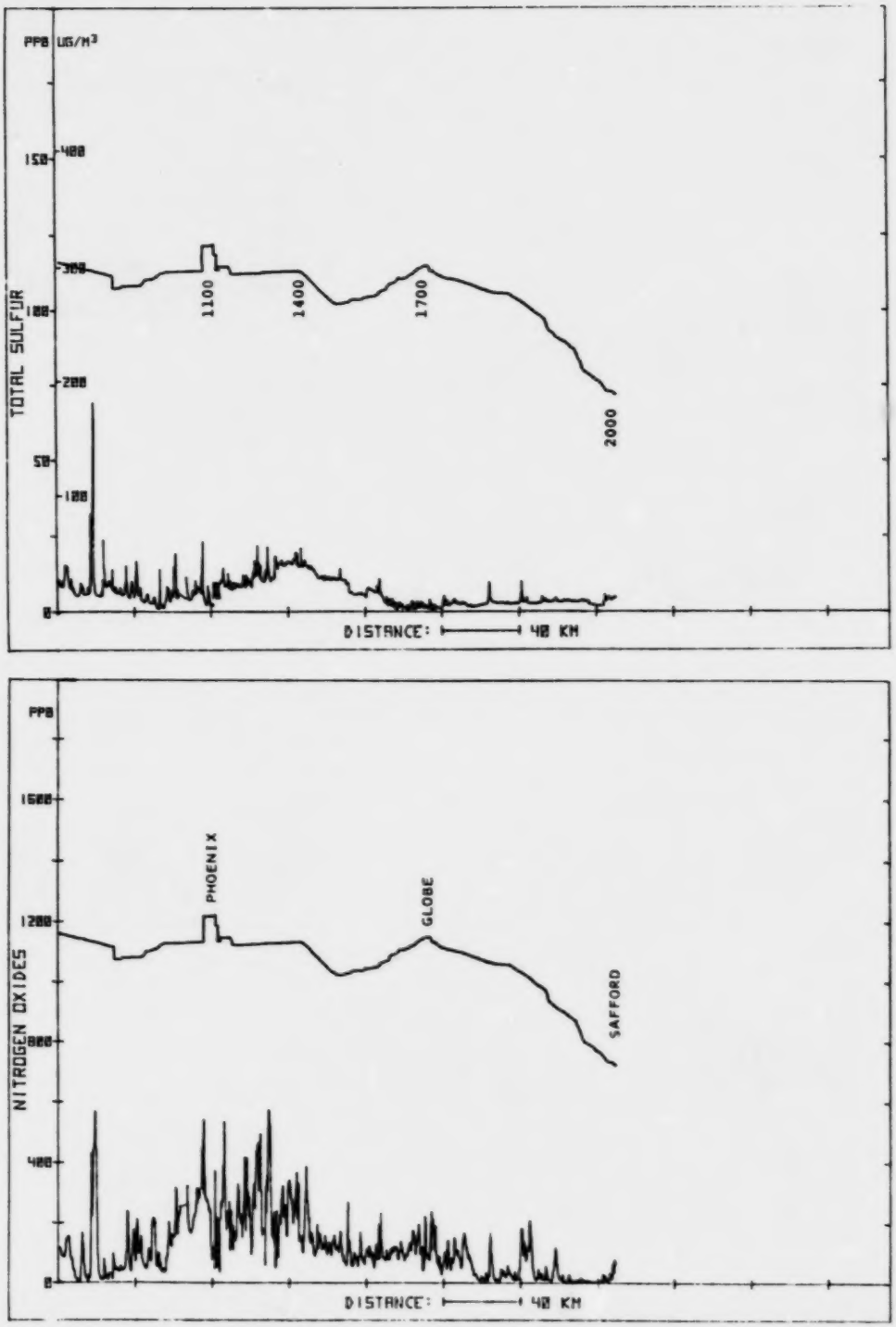

Figure 17. November 7 - 1100-2000 MST - Arizona

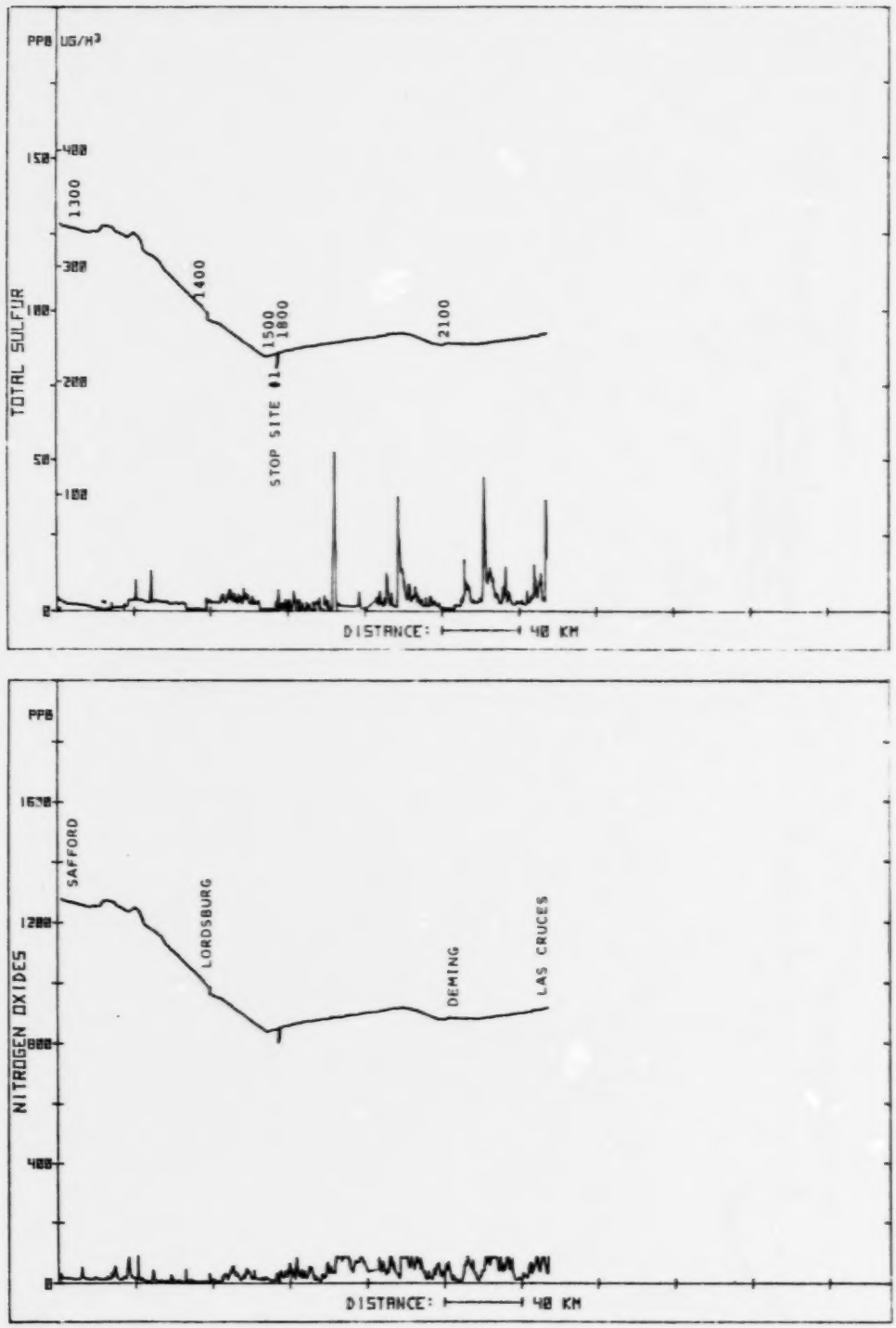

Figure 18. November 8 - 1300-2200 MST - Arizona-New Mexico

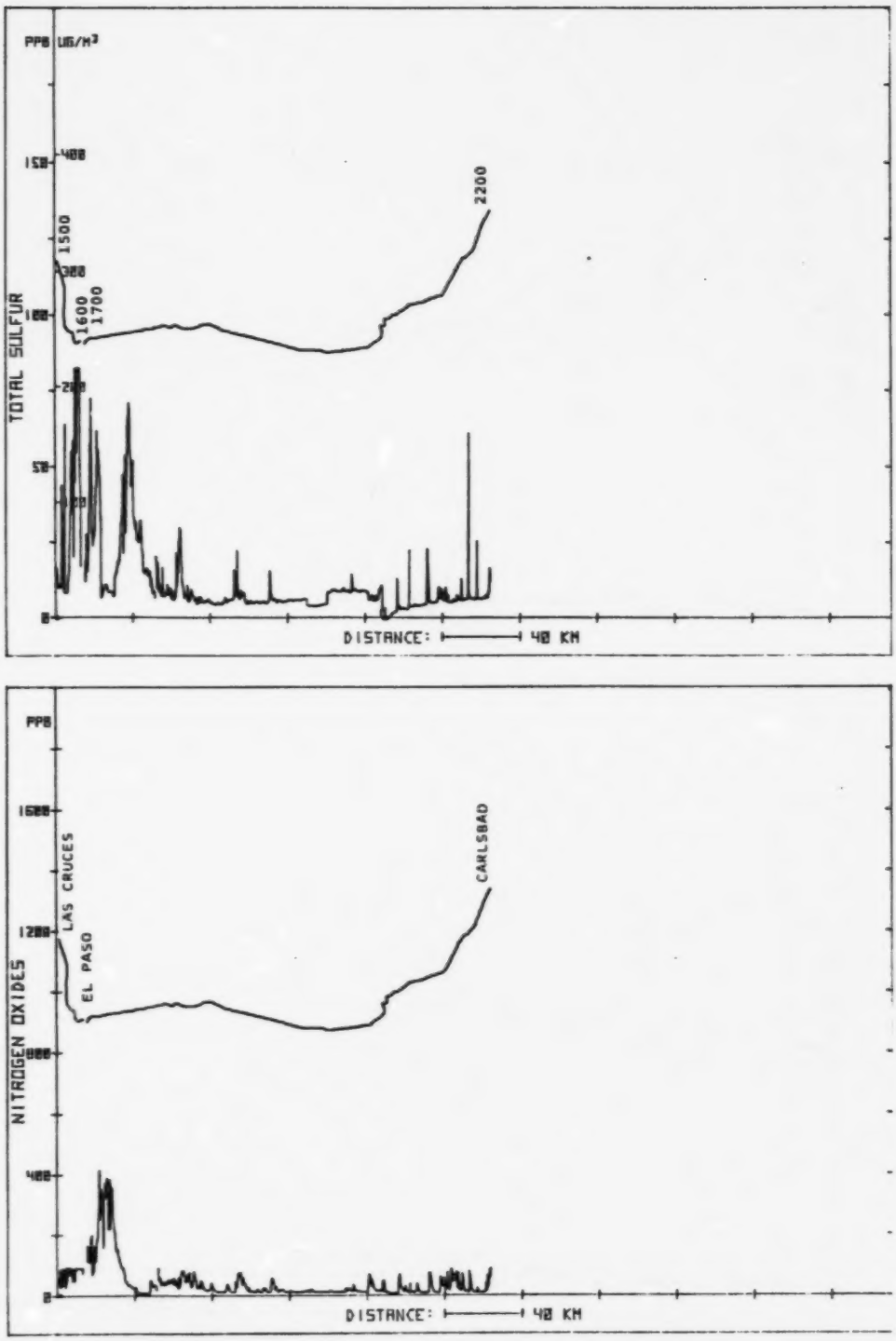

Figure 19. November 9 - 1500-2200 MST - New Mexico-Texas

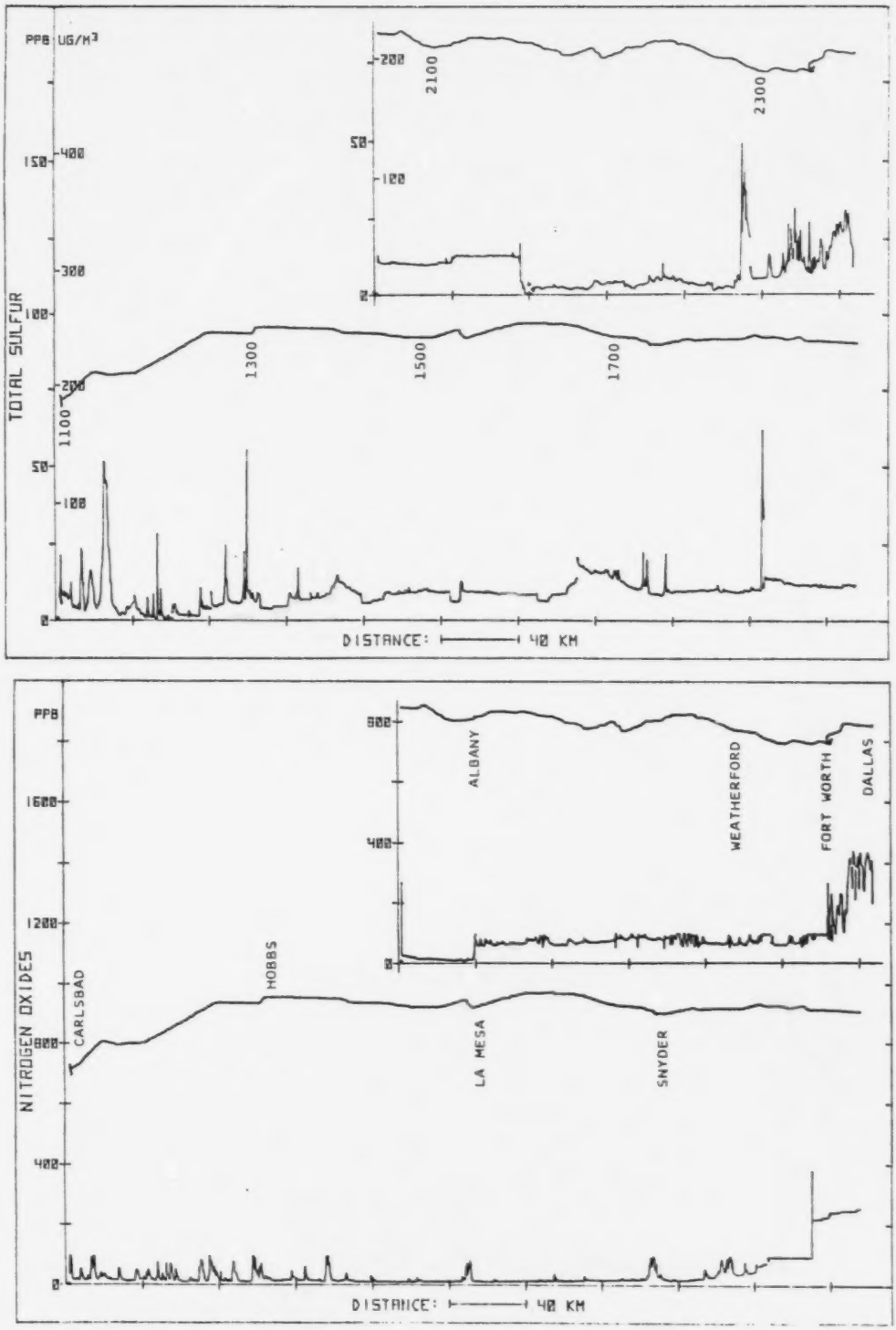

Figure 20. November 10 - 1100 MST to 2400 CST - Texas

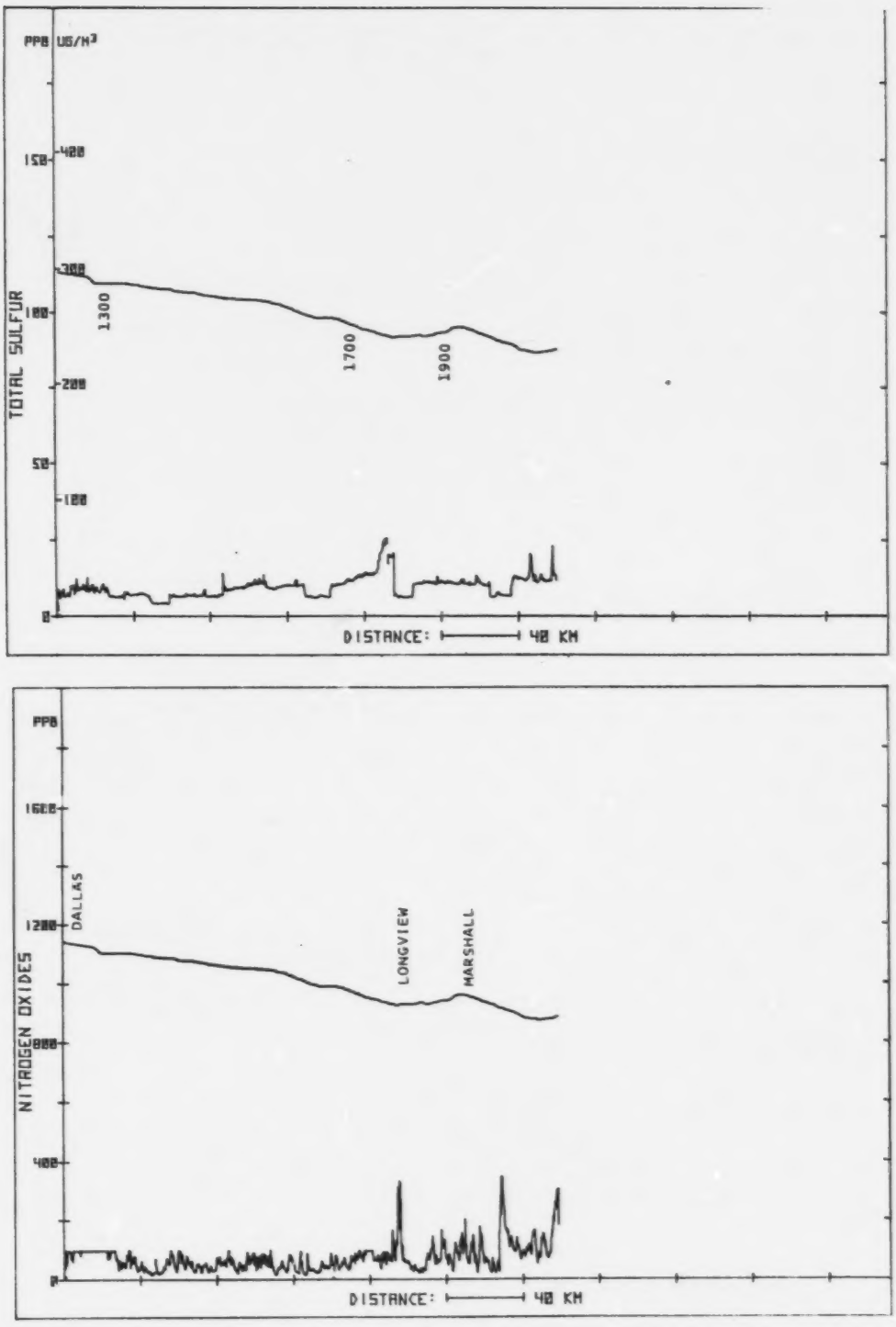

Figure 21. November 14 - 1200-2000 CST - Texas-Louisiana

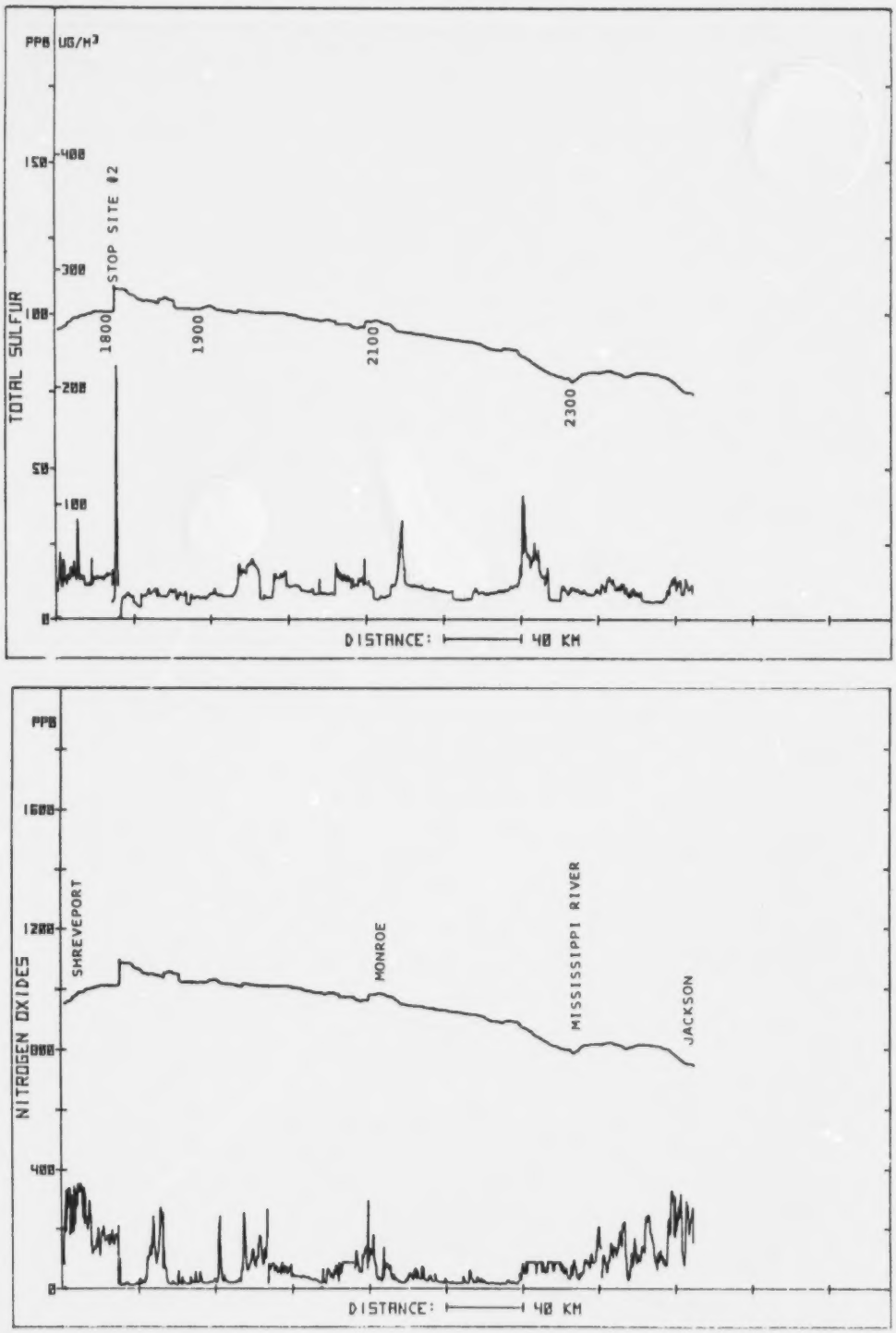

Figure 22. November 15 - 1400-2400 CST - Louisiana-Mississippi

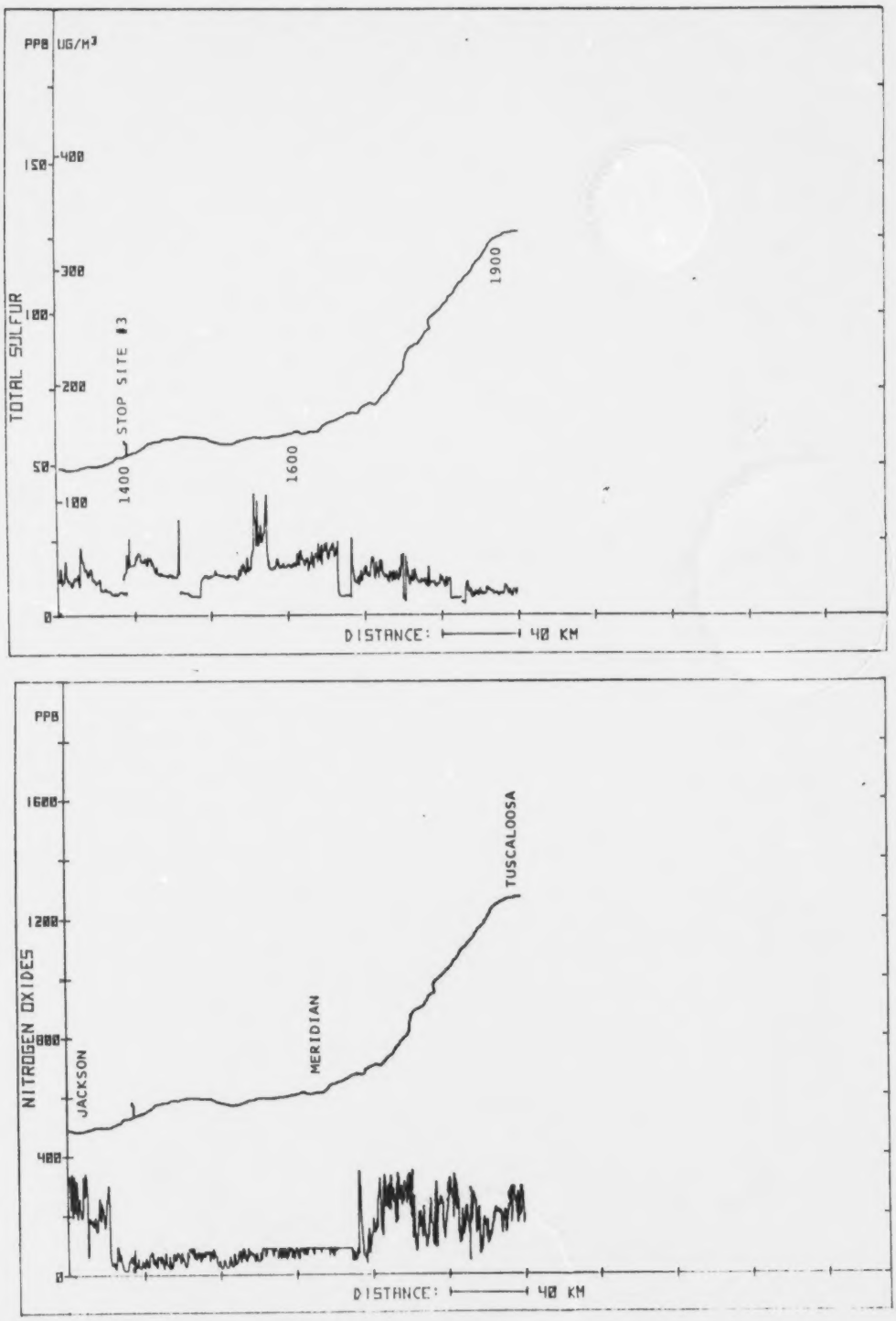

Figure 23. November 17 - 1000-1900 CST - Mississippi-Alabama

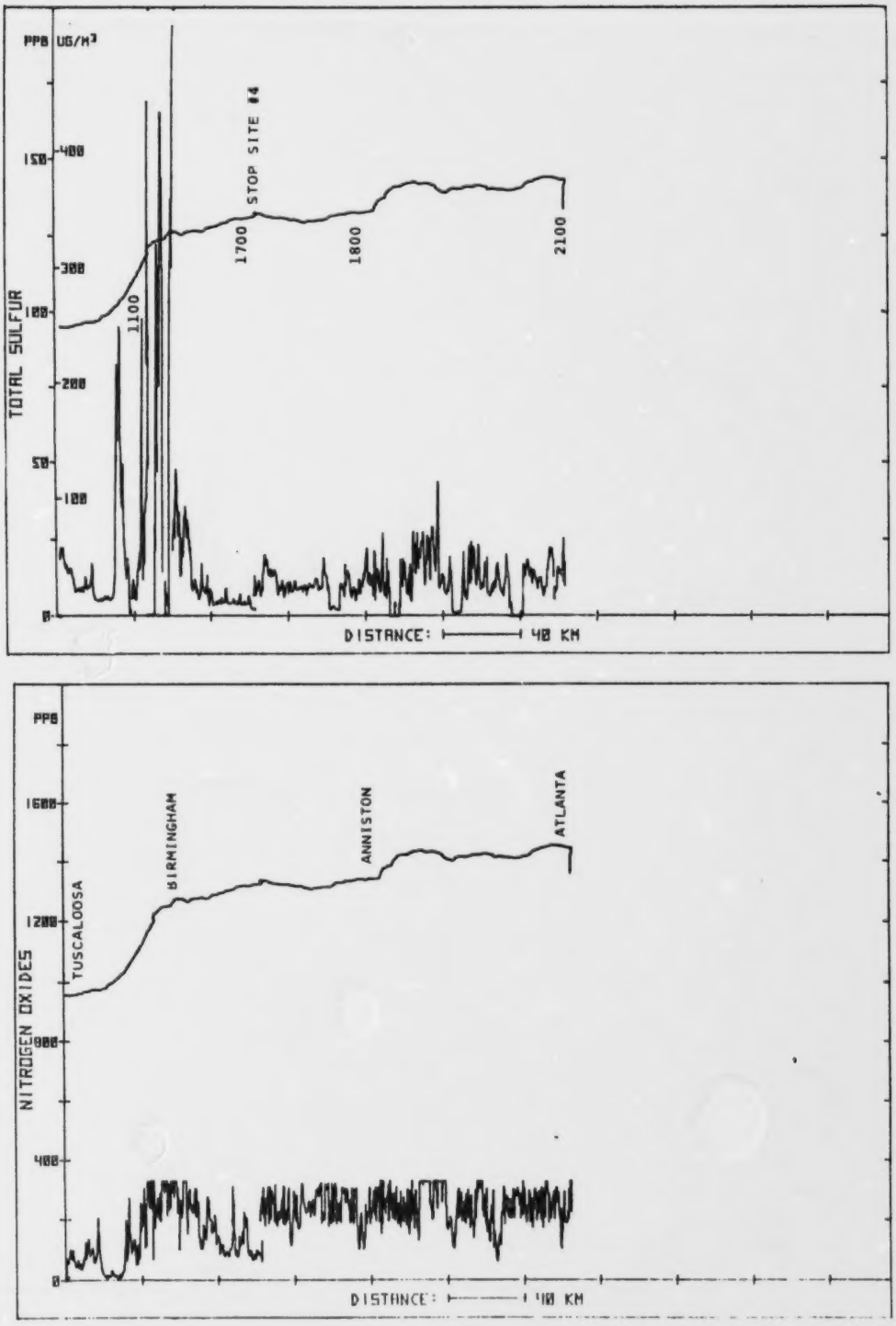

Figure 24. November 18 - 1000 CST to 2100 EST - Alabama-Georgia

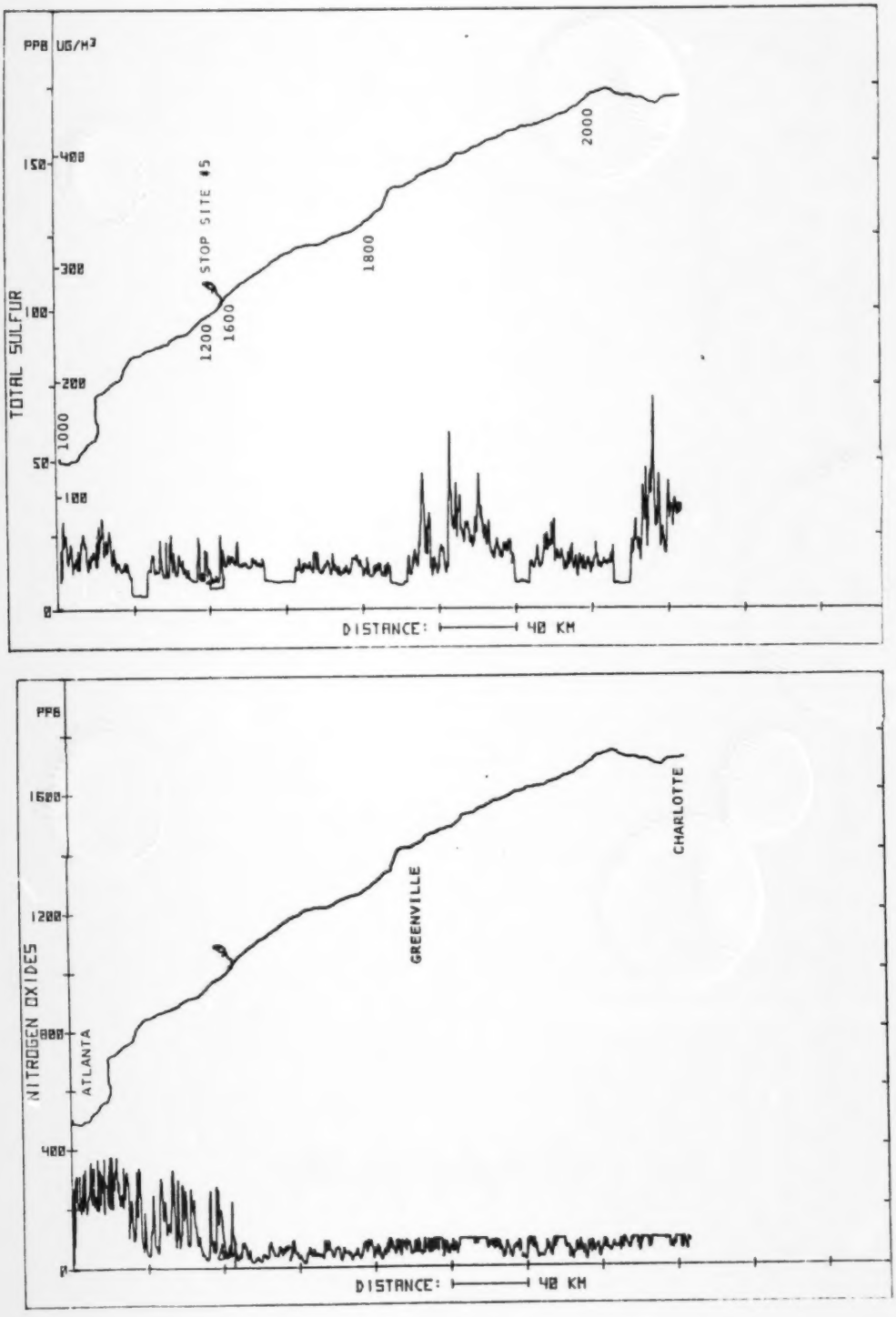

Figure 25. November 19 - 1000-2100 EST - Georgia-North Carolina

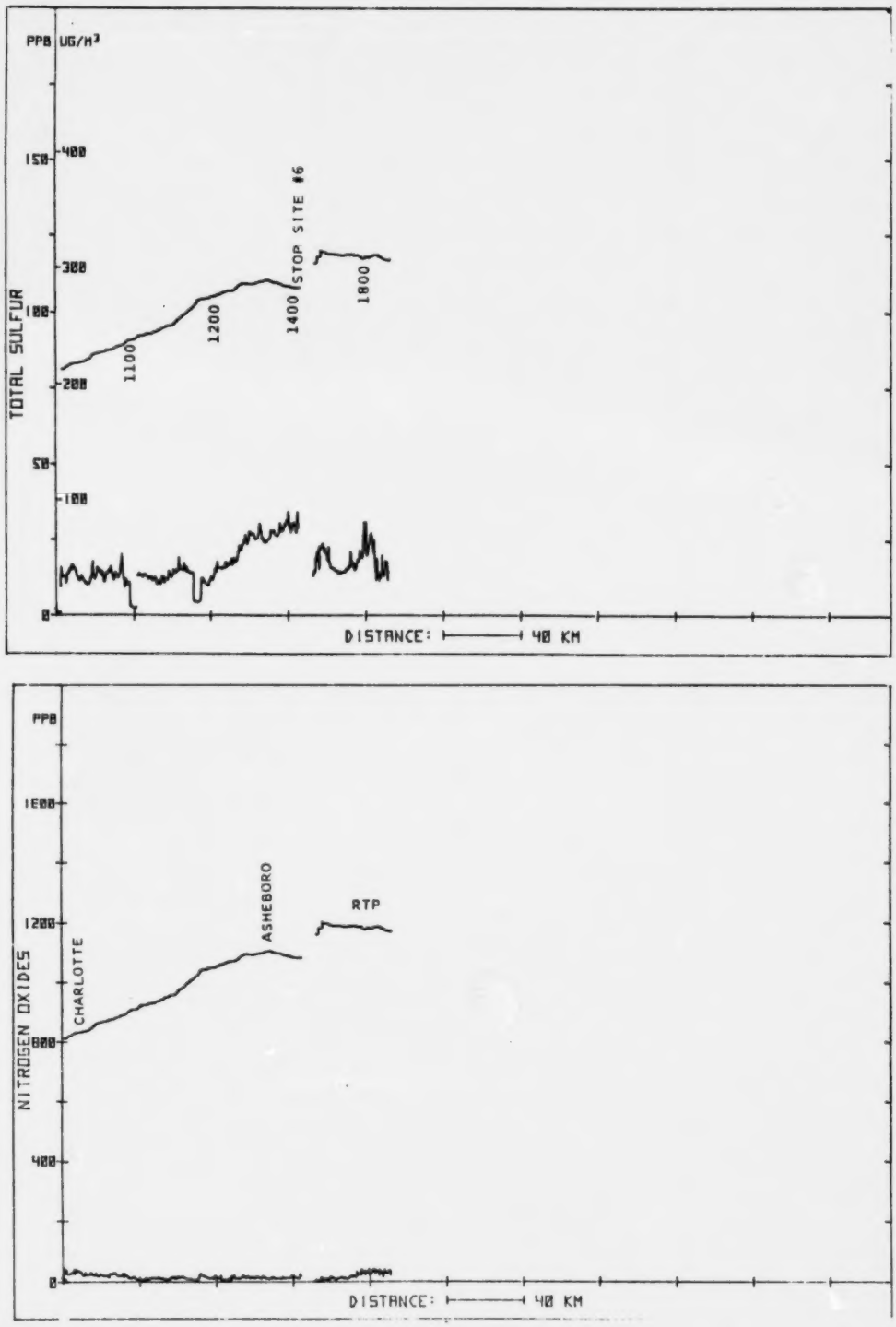

Figure 26. November 20 - 1000-1800 EST - North Carolina

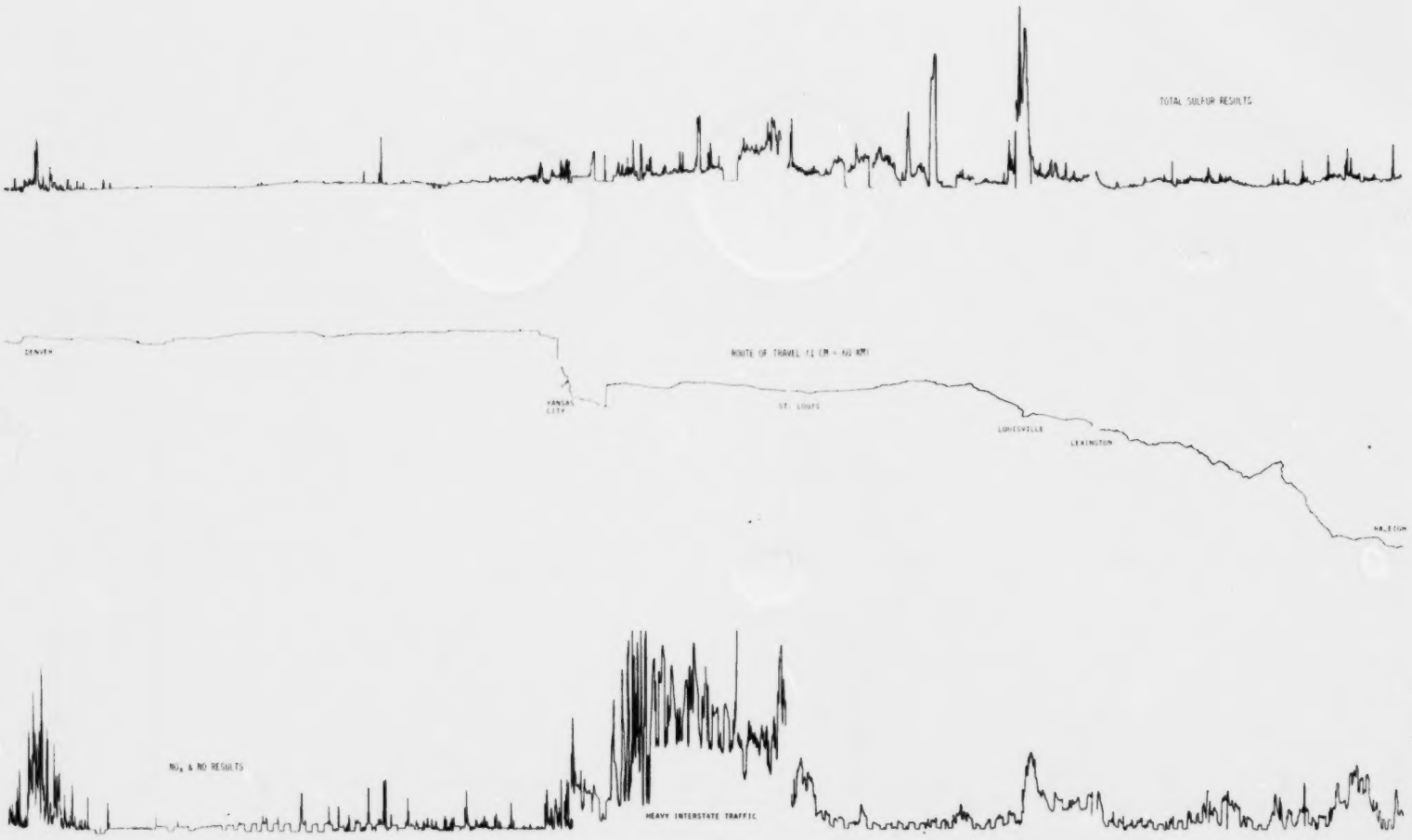

Figure 27. Trip 1 data summary - Denver to Raleigh - August 18-27, 1976

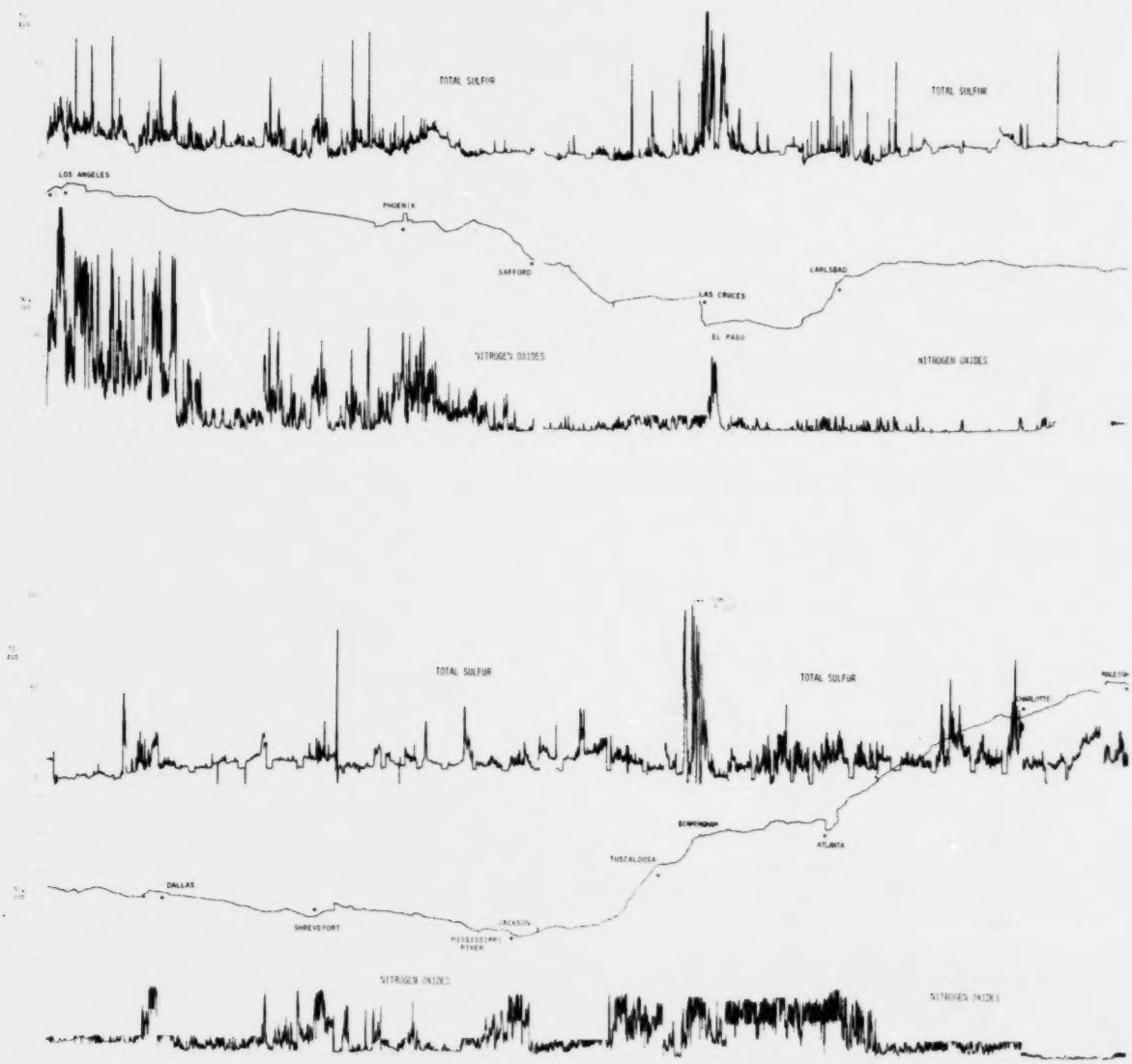

Figure 28. Trip 2 data summary - Los Angeles to Raleigh - November 4-20, 1976 (before data adjustments)

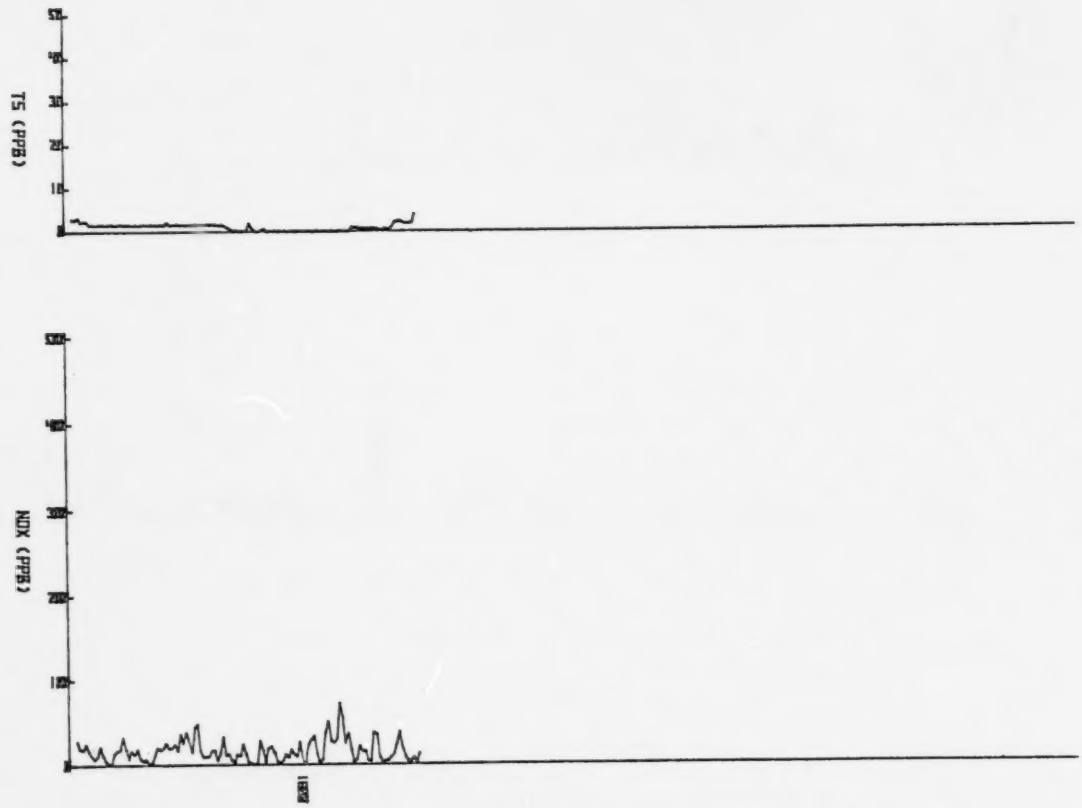

Figure 29. Stop site east of Phoenix (Superior, AZ) - November 7; stationery data only (no filter samples)

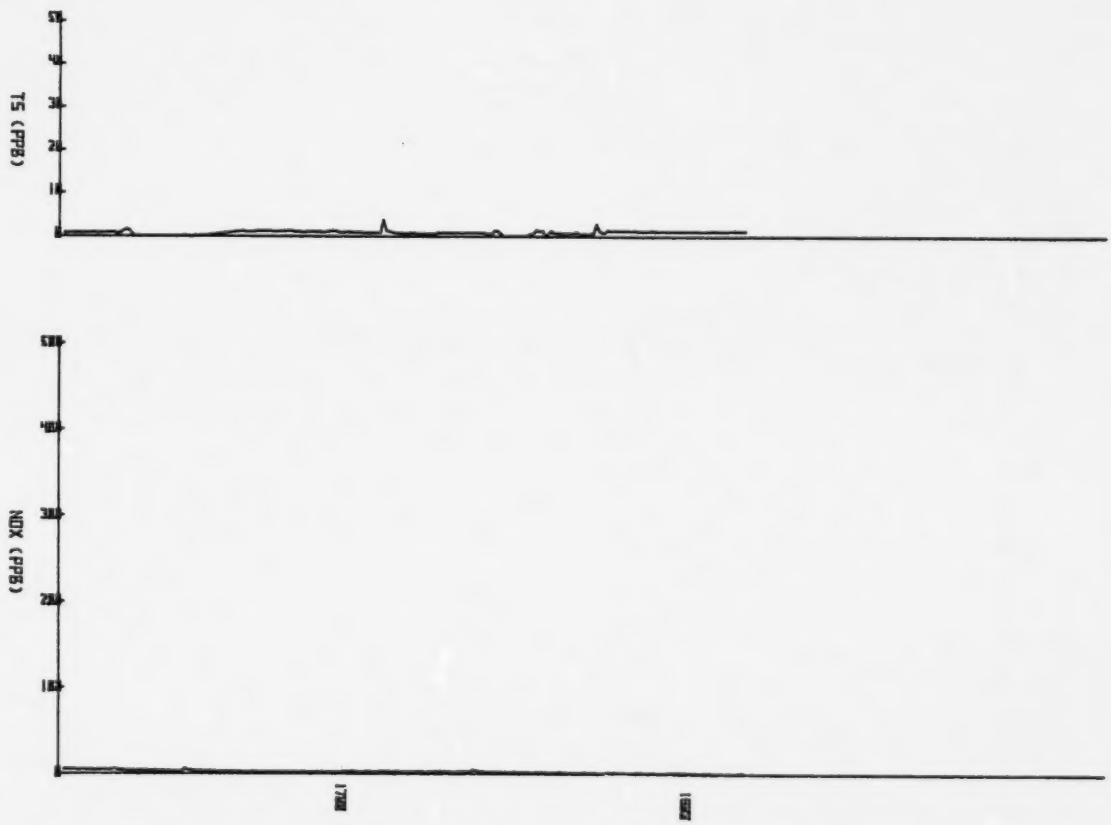

Figure 30. Stop site #1 - November 8 - Lordsburg, NM

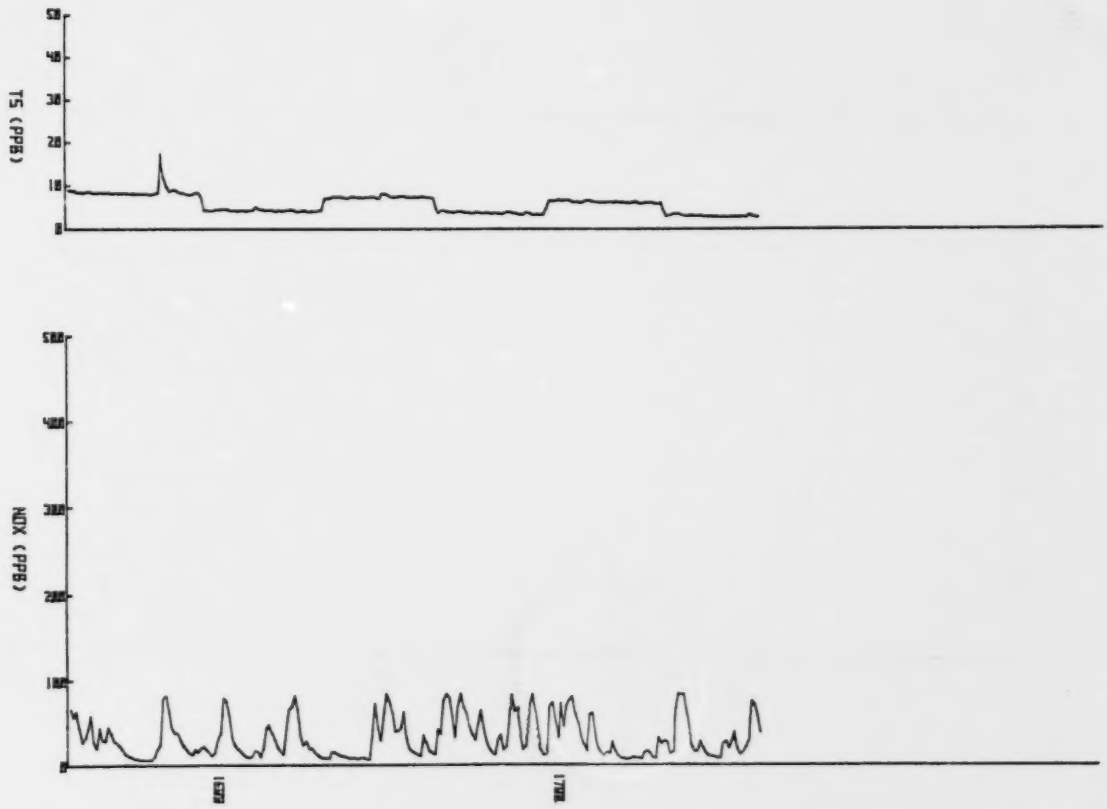

Figure 31. Stop site #2 - November 15 - Shreveport, LA

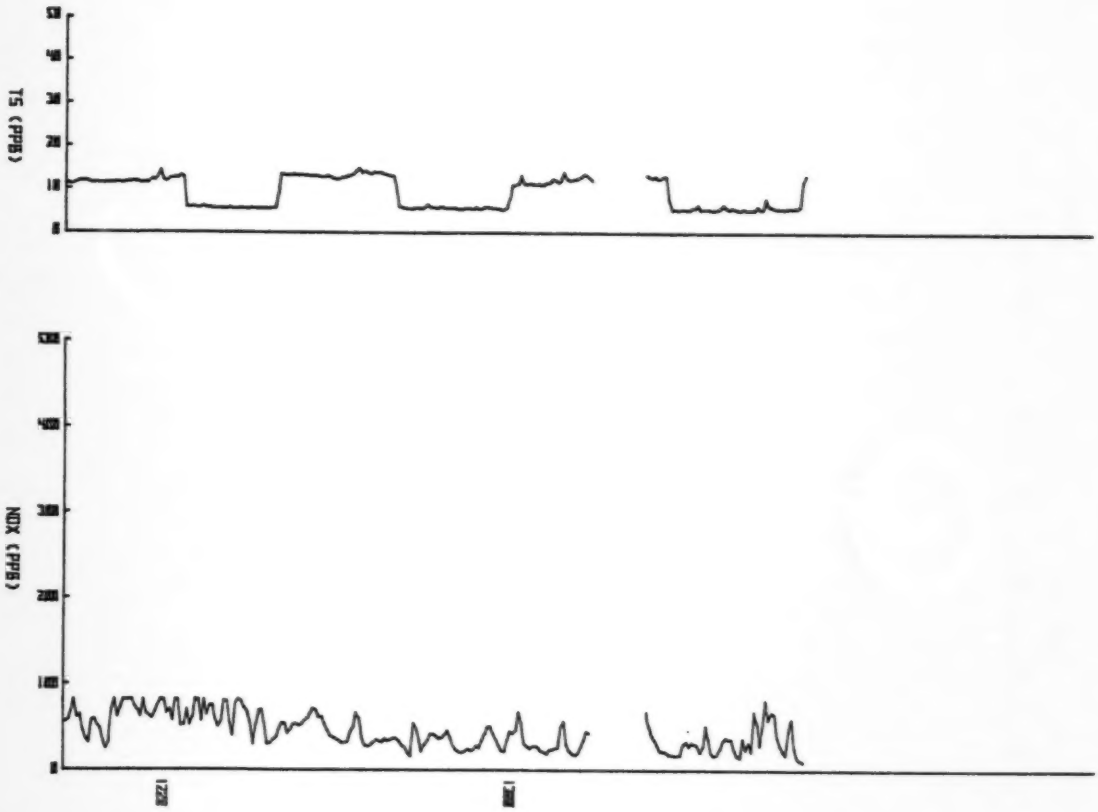

Figure 32. Stop site #3 - November 17 - Pelahatchie, MS

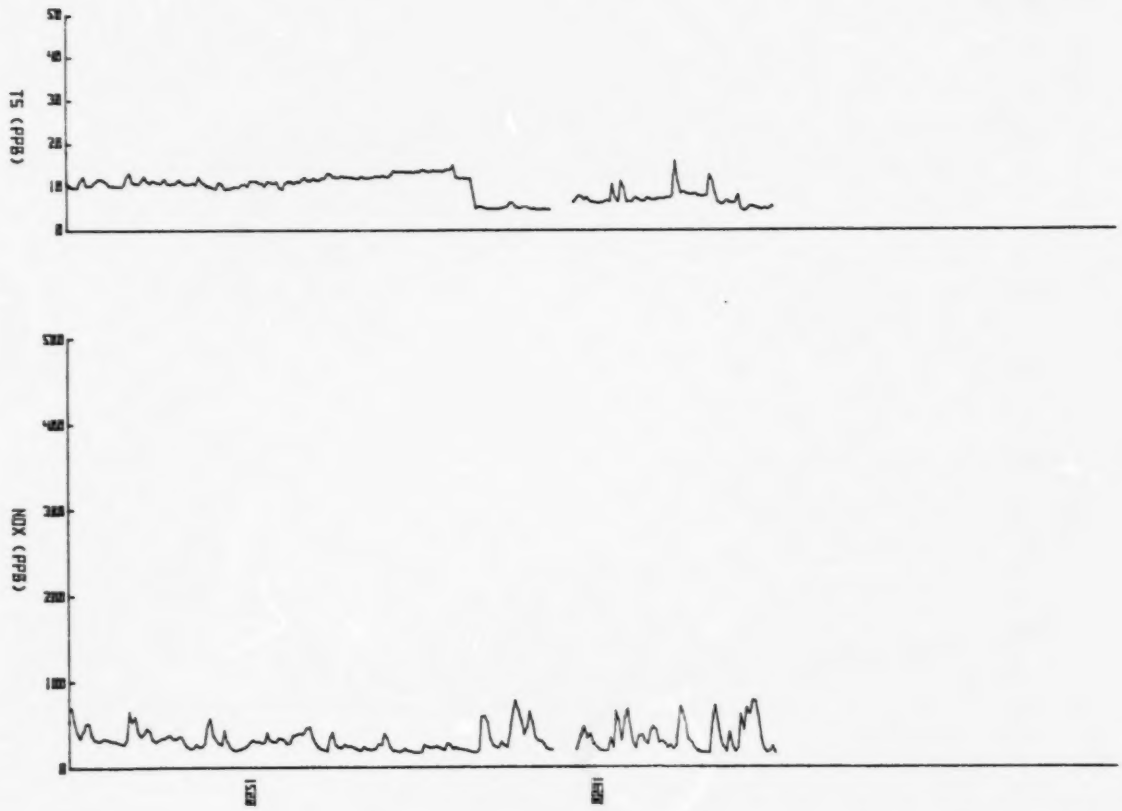

Figure 33. Stop site #4 - November 18 - Ragland, AL

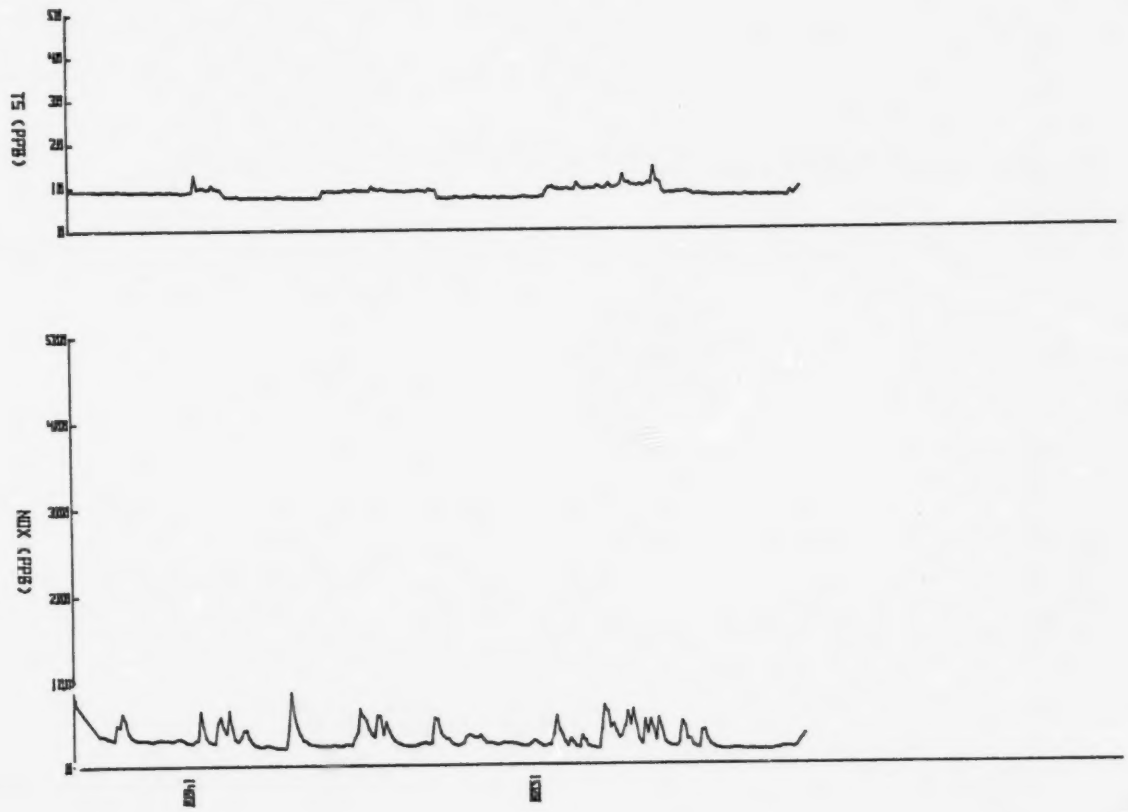

Figure 34. Stop site #5 - November 19 - Commerce, GA

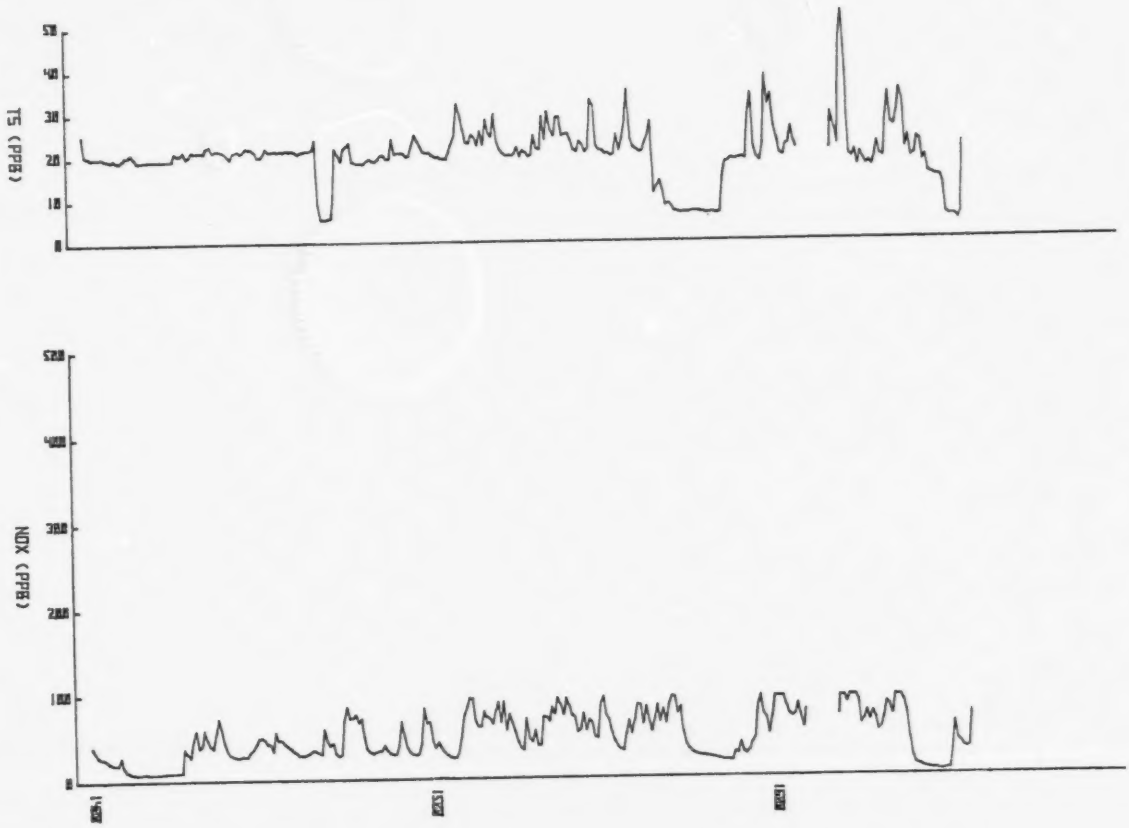

Figure 35. Stop site #6 - November 20 - Silver City, NC

SECTION 4

DISCUSSION

MOVING RESULTS

These results clearly show long distances of consistant quasi-uniform data between urban *islands* of higher content. Rural levels differed regionally; between some cities the levels were very low, while higher backgrounds existed between others.

Low levels of total sulfur, below one part per billion, were experienced; plumes of over a hundred ppb were crossed; tens of ppb were common in urban measurements. Higher rural backgrounds were present from the Mississippi River east.

In the nitrogen oxides' plot the switching to NO measurements during the August trip clearly results in troughs, which demonstrates that NO_X mainly consists of NO_2. During the November trip only NO_X was recorded.

There was a purposeful intent to travel on and off the Interstate Highway System along the route. The Interstate segments of travel resulted in much higher NO_X levels due to the traffic. The effect in the total sulfur results was less obvious and more closely related to individual vehicles. Diesel vehicles were very sulfurous. Travel in and for some distance east of Los Angeles reflected the content of air breathed by many automobile commuters. NO_X levels were normally in excess of 300-400 ppb.

Low levels of NO_X were observed to a few ppb in the western deserts. In the South, levels rose to tens of ppb; in urban areas and on highways levels exceeded hundreds of ppb frequently. Off heavily travelled streets, upwind of concentrated traffic, levels of several tens of ppb remained in urban locations.

STATIONARY MEASUREMENTS

At the seven stop sites the levels of sulfur were less than 10 ppb except at the last site, where levels of 50 ppb were reached. The NO_X measurements showed background levels of less than 25 ppb at all sites except Stop Site #6, which was near a dairy feed lot in a heavily agricultural region. The NO_X signal, which was often above 50 ppb during this final test, was almost entirely removed by the NH_3 denuder.

SECTION 5

ANALYSIS

After the cross-country trips, it was possible to reduce the data to a calibrated form and to plot them as a function of distance. A variety of plots were reviewed, and the preliminary analysis of the data, resulting from an overview of these plots, was presented in preliminary reports. Plots of this nature are found in Section 3. It was obvious that anomalies of pollution occurred in the vicinity of cities that were crossed during the trips and that these were separated by broad regions of considerably less pollution. A method of displaying an overview of these data in some uniform fashion was sought.

Initial attempts were to prepare plots of correlation vs the separation of data points. The initial assumption was that, over broad regions, adjacent and even widely separated data points would be similar for rural areas and dissimilar for urban areas. Therefore, one could provide an analysis to separate the character of the region traversed. This approach did not proceed well, primarily because most of the data points were near the noise level of the gaseous concentration being measured. One part per billion does not correlate very well with two parts per billion, and such threshold values were not uncommon in the western states. Another way was sought.

While preparing these materials, Environmental Measurements was visited by an associate from France, Dominique Rust from the Commissariat de Energie Atomique (CEA). He described a technique suggested by Dr. Pierre Zettwoog to provide a common format for similar long-distance traverses conducted by CEA. The technique is to provide a frequency distribution for the occurrence of a given concentration in a given set of data. Once this is calculated, a cumulative frequency distribution provides an integrated value that is distinctive for similar sets of data points. This method proved useful, as follows:

In the data that were accumulated for the two transcontinental trips, common dimensions have been used in Figures 36 through 50. The concentration for both nitrogen oxide and total sulfur is plotted as the log of concentration from one part per billion to one thousand parts per million on the abscissa. On the ordinate, the left edge is from 5-20% of the frequency occurrence of

the concentrations, which are plotted as dots. The right ordinate is 100% of the cumulative frequency distribution (integral curve), plotted as a solid line. If the cumulative plot is steep, the data tend to be contained within a narrow region. To the left they are of lower concentrations, and to the right they represent more significant pollution. A more gradual slope represents a broader distribution of the occurrence of the contaminants.

The presentations are ordered by showing the total sulfur data first and then the results from the nitrogen oxide measurements. In each case the results of the first trip are presented and followed by those of the second trip. Throughout both trips, while the data were gathered at 200-meter increments, each data point represents an average of ten samples. Thus, for example, the 19,026 points represented in the second trip's total sulfur presentation actually represents 190,260 samplings of the analyzers.

In each graph a location is identified, together with the number of data points used for the presentation. For each trip and for each gas the results of the entire trip are presented on a single page. The variation in the number of points used for each gas is a result of minor periods when one or another instrument may have been off during the course of the trip. The "Entire Trip" results are followed by subsets in urban areas traversed by the trip and by subsets in regional rural traverses between cities. Each takes a distinctive form. No attempt was made to modify the data that were recorded on the trip other than to reduce them to engineering units. The spikes that occur as a result of traffic are thus intermixed with the results of transecting the touchdown of plumes or the general air throughout the cities on the route. Periodic daily calibrations and spans are also not excised from the data, but they represent a relatively small portion of the total data points and should not bias the results in any significant fashion. To a first order approximation, these summary data represent the air breathed by the occupants of the vehicle as it traversed across the United States.

TOTAL SULFUR RESULTS

An overview of Trip 1 versus Trip 2 would indicate that the results of both trips are similar with 90% of the measurements made falling beneath approximately 11 ppb total sulfur.

Selected urban sites show quite distinct differences, however. In Trip 1, for example, 90% of the total sulfur results in Denver fall below 8 ppb; yet in Louisville, this cut-off is approximately 100 ppb. This same higher level represents Birmingham, Alabama, from the second trip. The results from Phoenix and Los Angeles show somewhat more sulfur than do those from Denver

(90% below 12 ppb); these appear to be related to automotive traffic.

Rural results are also rather similar. The Great Plains trip between Denver and Kansas City is comparable to the desert trip between Phoenix and El Paso, with very little presence of total sulfur. The trip across the Allegeny Mountains from Lexington to Raleigh shows distinctly more total sulfur presence; the trip from Dallas to Birmingham across the southern Mississippi River valley shows a higher level and distinctly more presence of total sulfur. The time of the Lexington-Raleigh traverse was August 26-27, 1976, during the occurrence of a persistent elevated pollution episode, as defined by later analysis of regional data.

One must keep in mind that these data are presented as a preliminary analysis of the total results in an effort to present a means by which the regional data can be quantatively compared. They represent results as they occurred over randomly chosen times of travel; thus they may be assumed to be good approximations of what one may find at any time.

NITROGEN OXIDES

The results of the nitrogen oxide measurements are more difficult to evaluate because they were measured on each trip with different instruments and because on the first trip there was a periodic switching between the measurement of total nitrogen oxides and NO. The second trip measured only total nitrogen oxide measurements. Whereas it would be highly desirous to separate these data, this has not been done because the data were not gathered in a fashion to make this analysis convenient. The results of Trip 1 represent lower concentrations because of the values obtained when measuring NO only.

On Trip 2 one of the instruments that was used had a range position, which, when the measurements exceeded this range, rested at a limit. The fact that this was not monitored at all times is obvious in the results of the data shown in the entire trip. The large peak of over 3% occurrence at about 90 ppb represent this limit; these data should be spread to higher concentrations.

It must be remembered that frequent routes on both these trips are on Interstate highways and that there are frequent occurrences of automobiles' and other vehicles' proceeding just in front of the measuring vehicle. Spikes of measurements occur throughout the trips, but these spikes have little correlation. They were left in the data, again to represent the air that the operators and drivers were breathing.

With these limitations in mind, the summary representations still retain an intriguing overview of these cross-country trips.

As in the total sulfur data, the NO_X data for both entire trips are rather similar, with 90% of the results occuring in less than 200 ppb. The results from the urban measurements in Denver, Louisville, and Phoenix were all rather similar, with 90% of the occurrences falling less than approximately 300 ppb. Los Angeles met expectations with the higher occurrence of NO_X, and Birmingham, while notable in total sulfur content, was low in the presence of NO_X. In the rural results each selected regional traverse had relatively little traffic. The trip between Denver and Kansas City was on a little-travelled U.S. highway, and the peaks and troughs of the frequency occurrence resulted from the switching from the NO to NO_X. Levels of less than approximately 40 ppb NO_X were present for 90% of that trip. This compares favorably with the relatively untravelled route between Dallas and Birmingham. Results between Lexington and Raleigh represent a more heavily travelled route and slower transit times through the mountains. These compare with the data gathered between Phoenix and El Paso; yet it must be observed that there were *very* few vehicles on the western route, and the results may represent a more regional presence of NO_X.

One is reminded that the entire purpose of both of these trips was to present an experimental means of gathering regional data that might be representative. In that same light, the results here are presented to suggest a convenient form for evaluation.

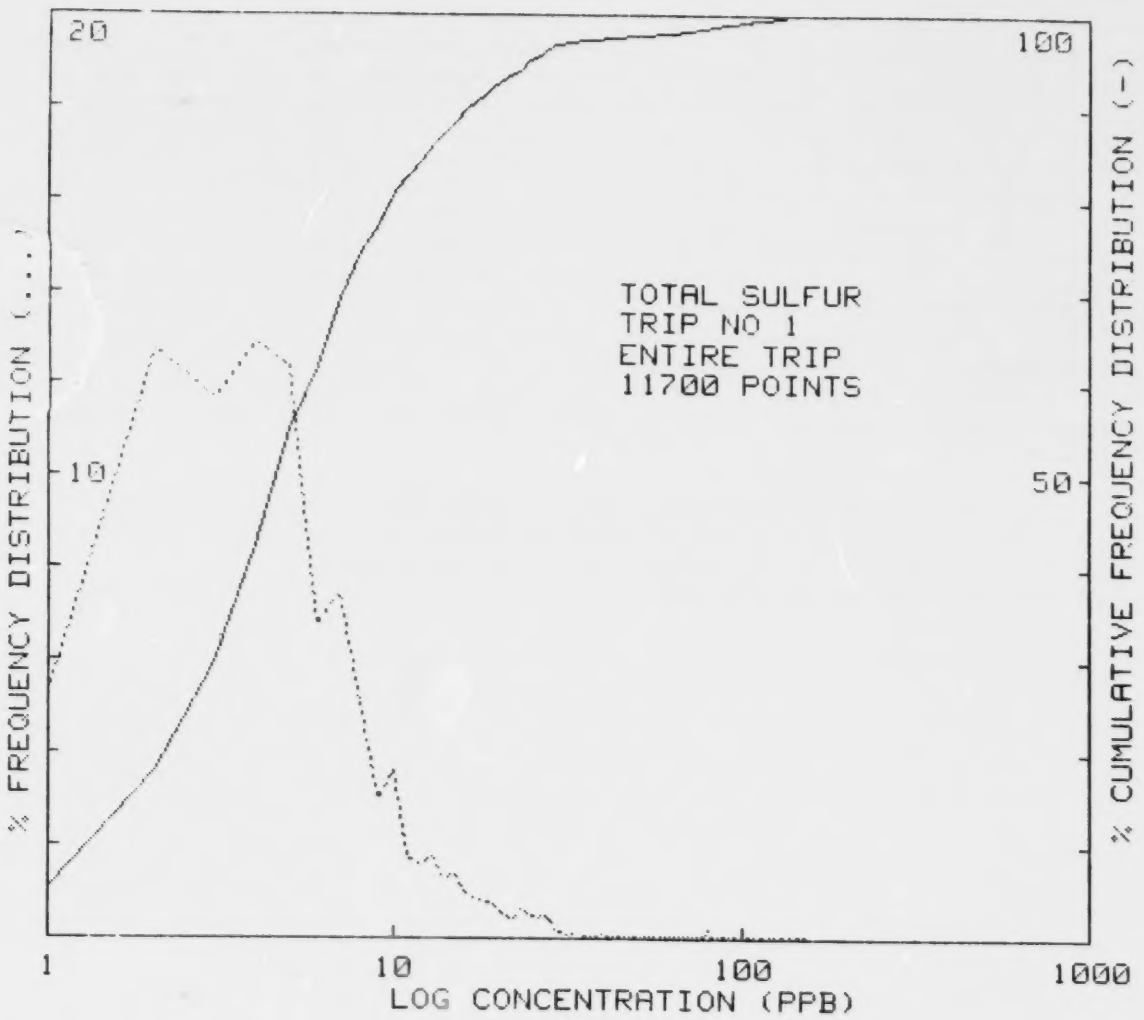
20
10
100
50
TOTAL SULFUR
TRIP NO 1
ENTIRE TRIP
11700 POINTS
% FREQUENCY DISTRIBUTION (...)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
1
10
100
1000
LOG CONCENTRATION (PPB)

Figure 36.

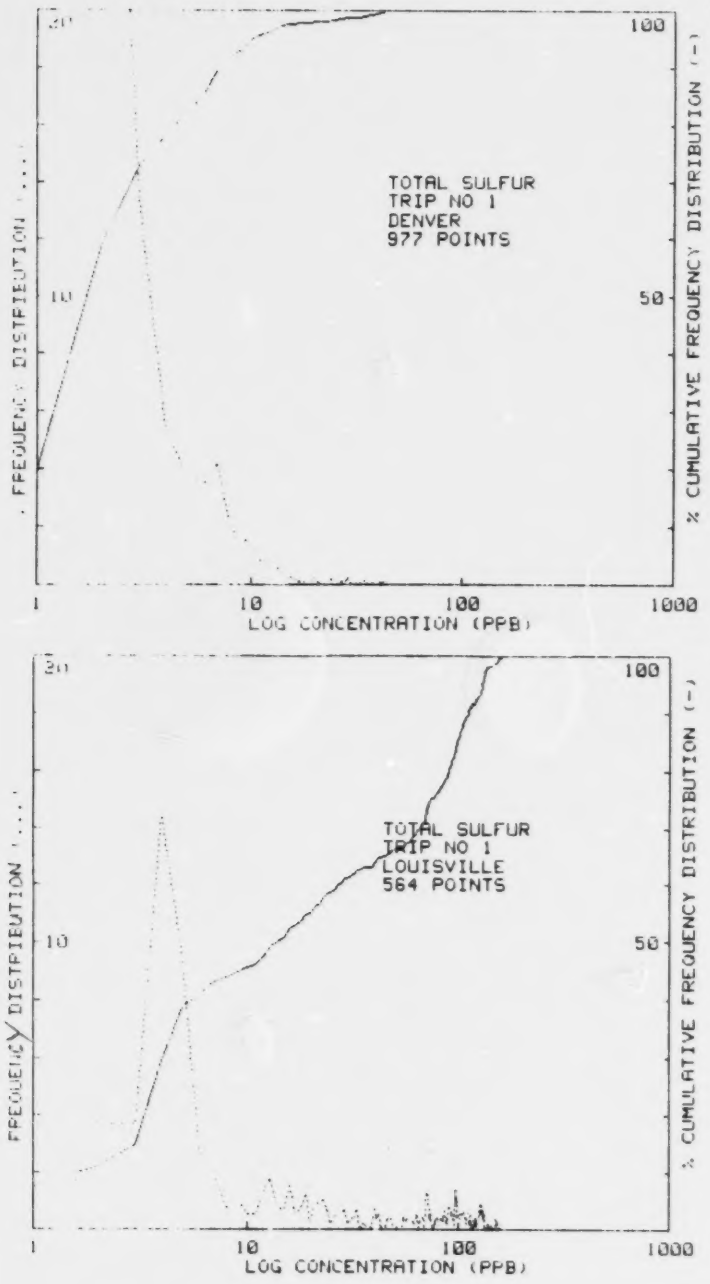
TOTAL SULFUR
TRIP NO 1
DENVER
977 POINTS
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
LOG CONCENTRATION (PPB)
1
10
100
1000
50
100
TOTAL SULFUR
TRIP NO 1
LOUISVILLE
564 POINTS
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
LOG CONCENTRATION (PPB)

Figure 37.

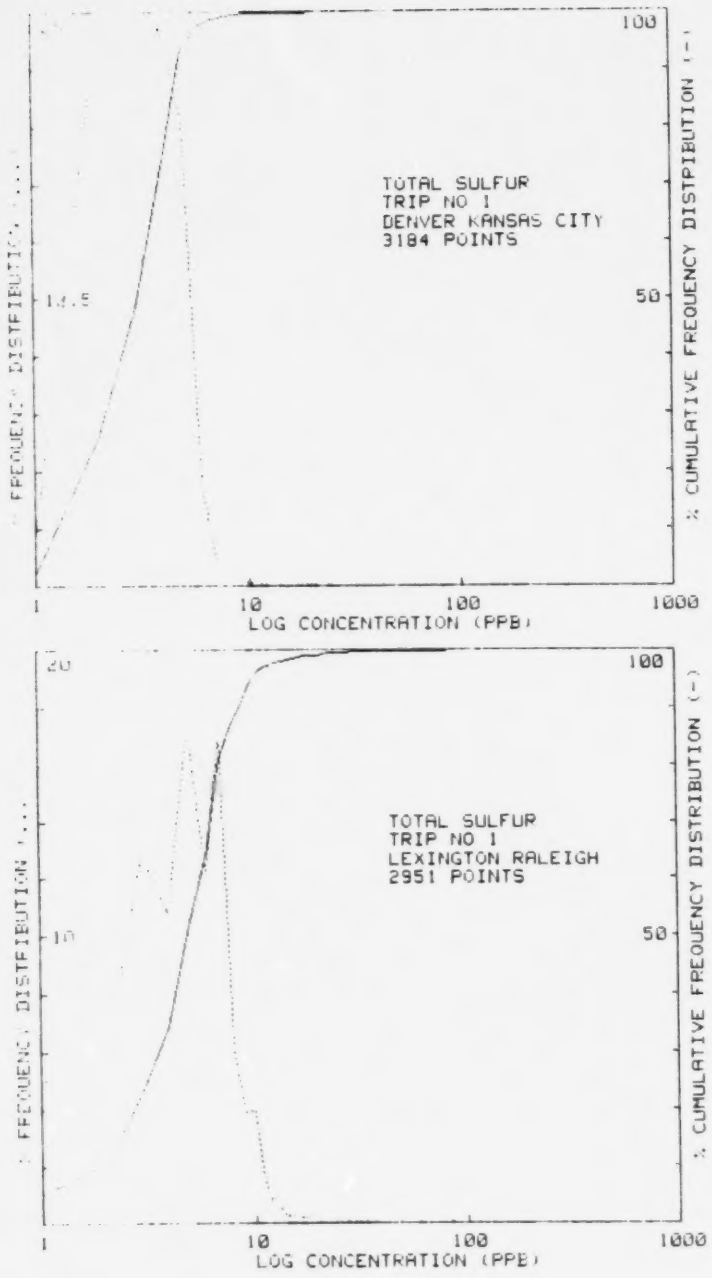
100
50
TOTAL SULFUR
TRIP NO 1
DENVER KANSAS CITY
3184 POINTS
1
10
100
1000
LOG CONCENTRATION (PPB)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
20
10
100
50
TOTAL SULFUR
TRIP NO 1
LEXINGTON RALEIGH
2951 POINTS
1
10
100
1000
LOG CONCENTRATION (PPB)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)

Figure 38.

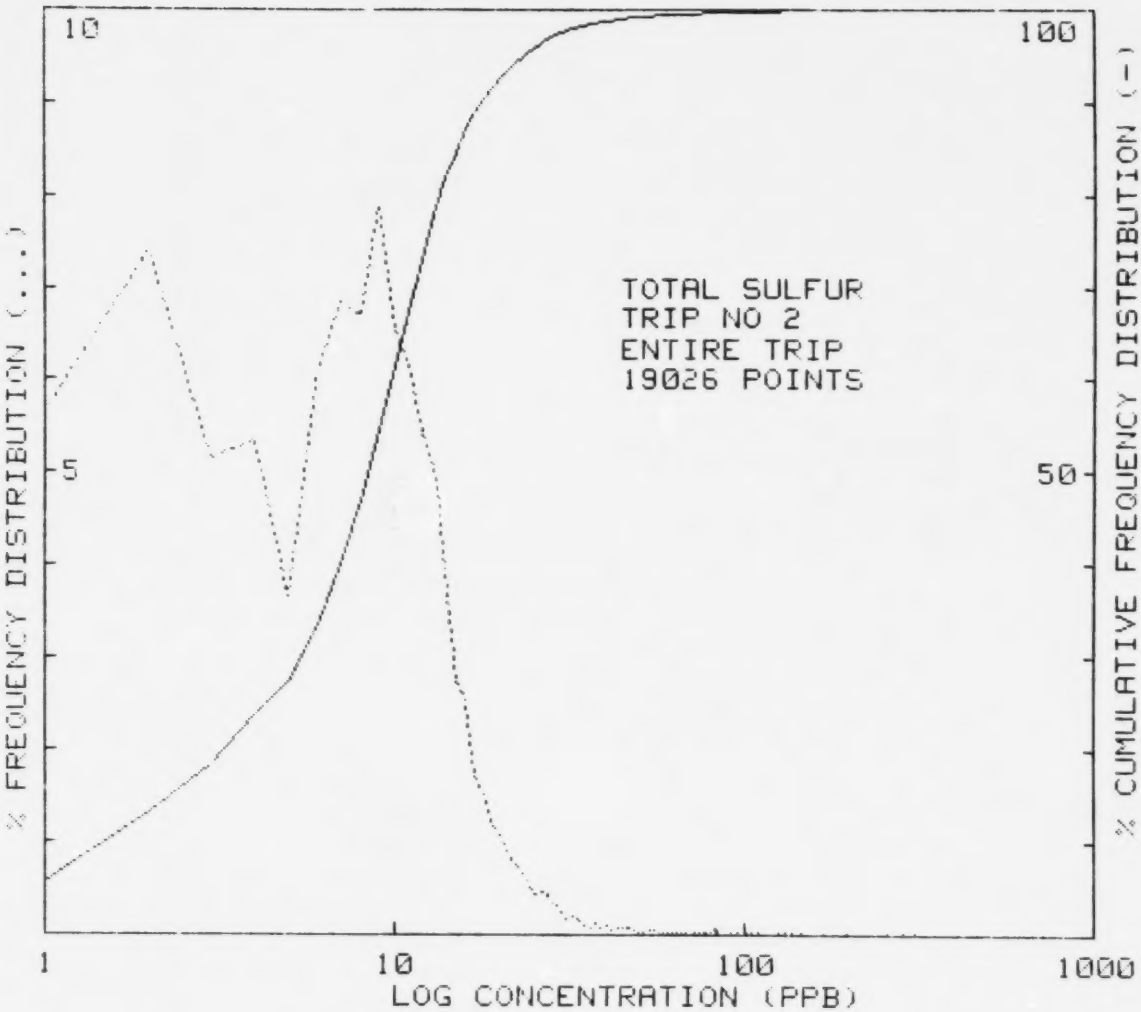
10
100
5
50
TOTAL SULFUR
TRIP NO 2
ENTIRE TRIP
19026 POINTS
% FREQUENCY DISTRIBUTION (...)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
1
10
100
1000
LOG CONCENTRATION (PPB)

Figure 39.

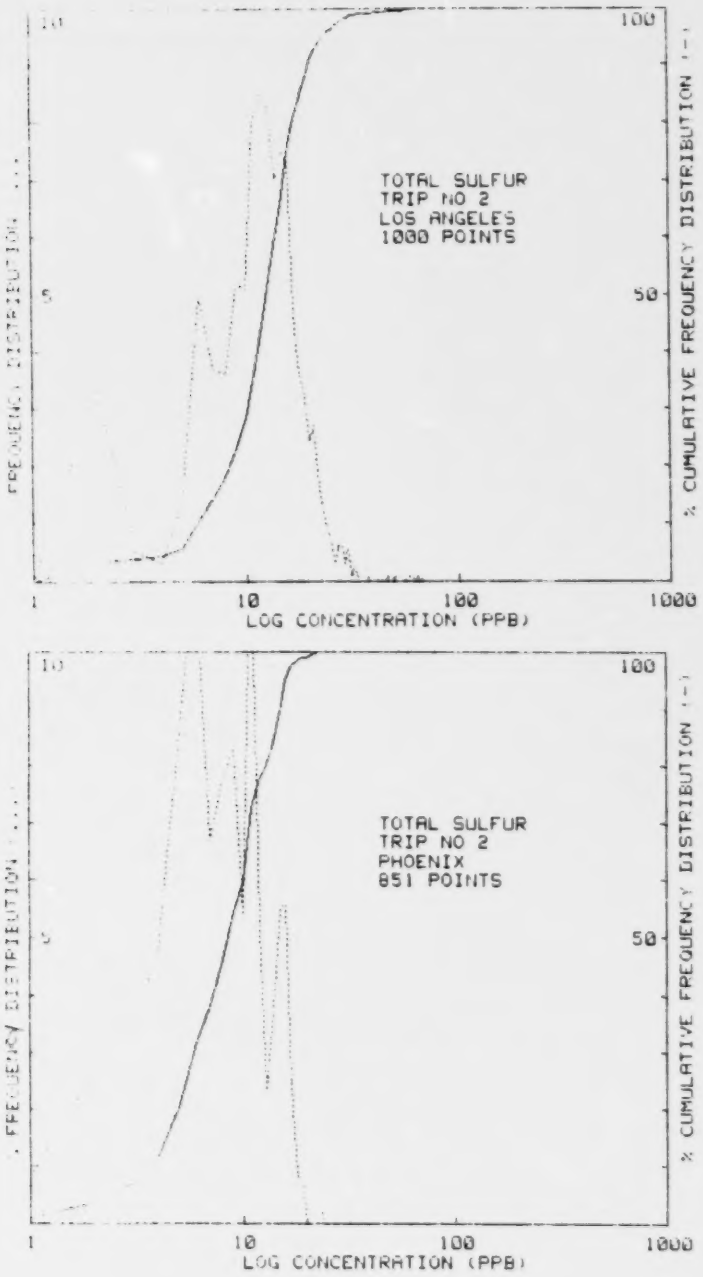
TOTAL SULFUR
TRIP NO 2
LOS ANGELES
1000 POINTS
100
50
1
10
100
1000
LOG CONCENTRATION (PPB)
% CUMULATIVE FREQUENCY DISTRIBUTION
TOTAL SULFUR
TRIP NO 2
PHOENIX
851 POINTS
100
50
1
10
100
1000
LOG CONCENTRATION (PPB)
% CUMULATIVE FREQUENCY DISTRIBUTION

Figure 40.

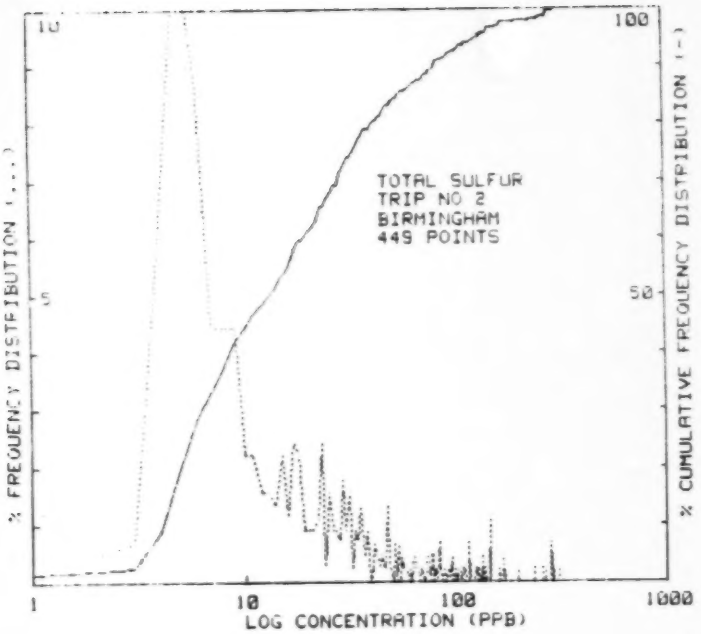
10
100
TOTAL SULFUR
TRIP NO 2
BIRMINGHAM
449 POINTS
5
50
% FREQUENCY DISTRIBUTION (...)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
1
10
100
1000
LOG CONCENTRATION (PPB)

Figure 41.

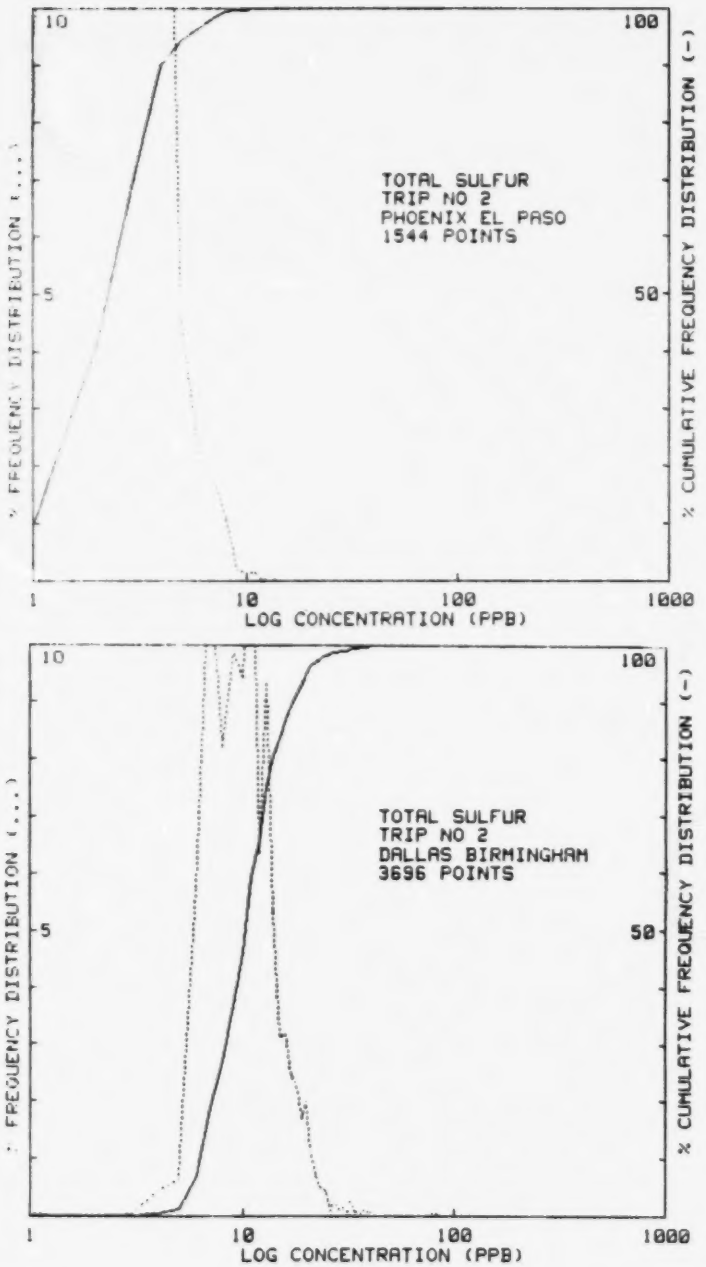
10
100
5
50
TOTAL SULFUR
TRIP NO 2
PHOENIX EL PASO
1544 POINTS
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
1
10
100
1000
LOG CONCENTRATION (PPB)
10
100
5
50
TOTAL SULFUR
TRIP NO 2
DALLAS BIRMINGHAM
3696 POINTS
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
1
10
100
1000
LOG CONCENTRATION (PPB)

Figure 42.

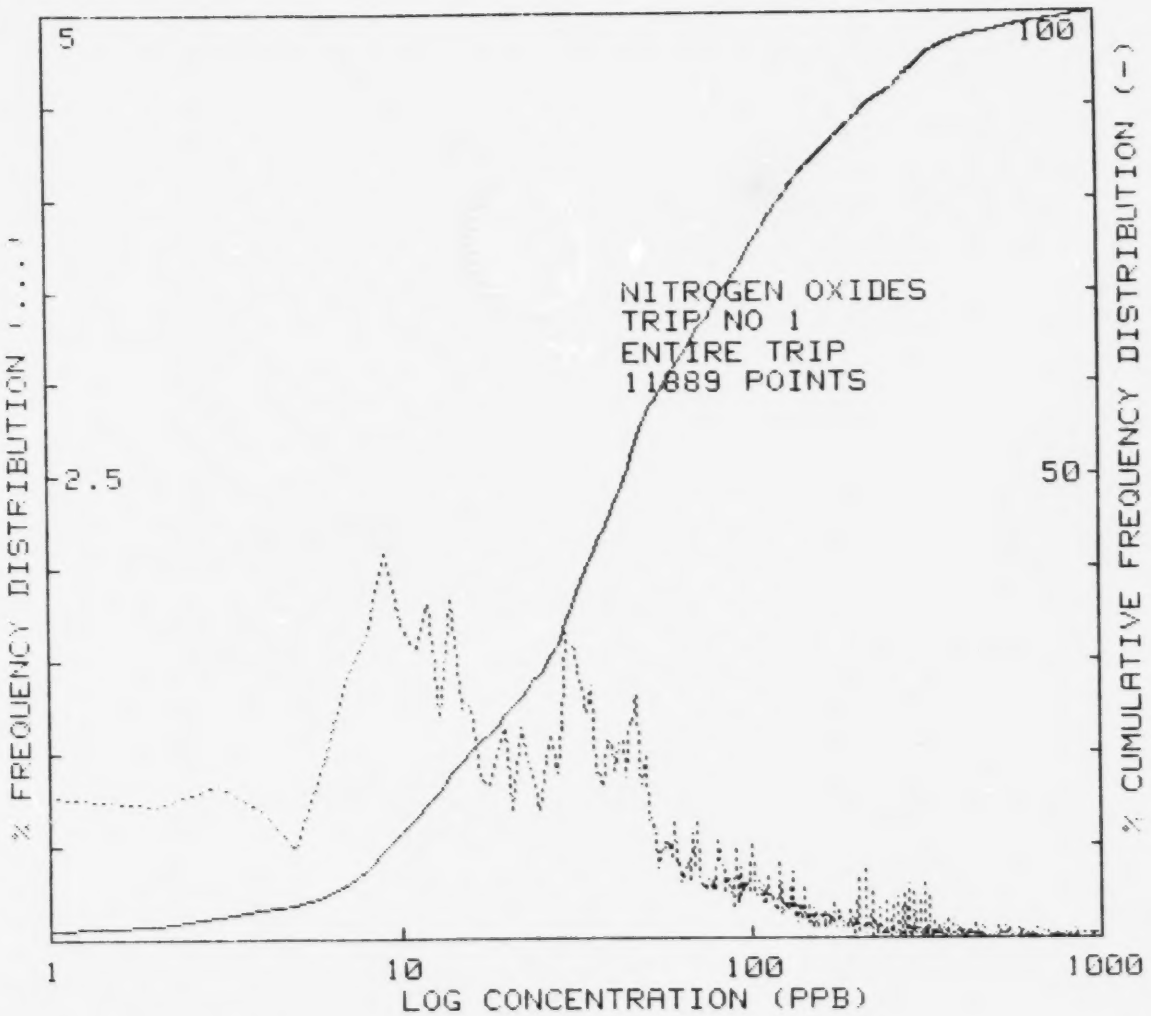
5
2.5
100
50
NITROGEN OXIDES
TRIP NO 1
ENTIRE TRIP
11889 POINTS
% FREQUENCY DISTRIBUTION (...)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
1
10
100
1000
LOG CONCENTRATION (PPB)

Figure 43.

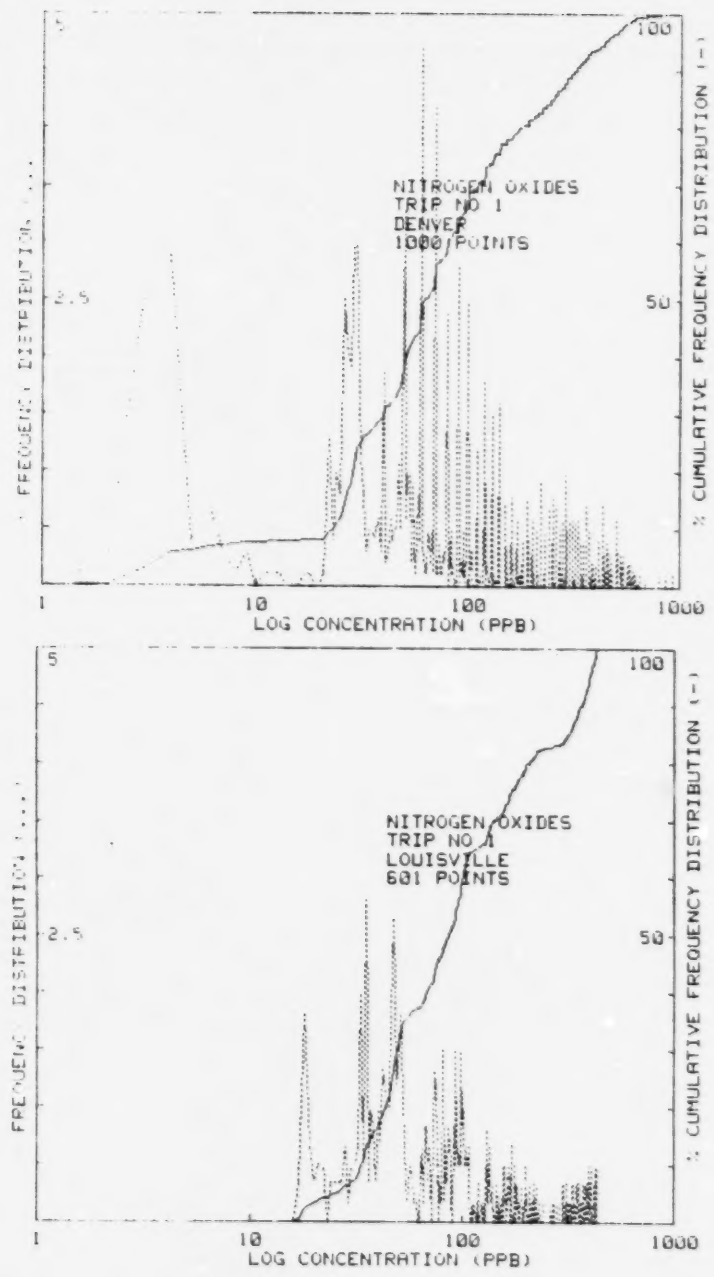
NITROGEN OXIDES
TRIP NO 1
DENVER
1000 POINTS
100
50
2.5
5
1
10
100
1000
LOG CONCENTRATION (PPB)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
NITROGEN OXIDES
TRIP NO 1
LOUISVILLE
601 POINTS
100
50
2.5
5
1
10
100
1000
LOG CONCENTRATION (PPB)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)

Figure 44.

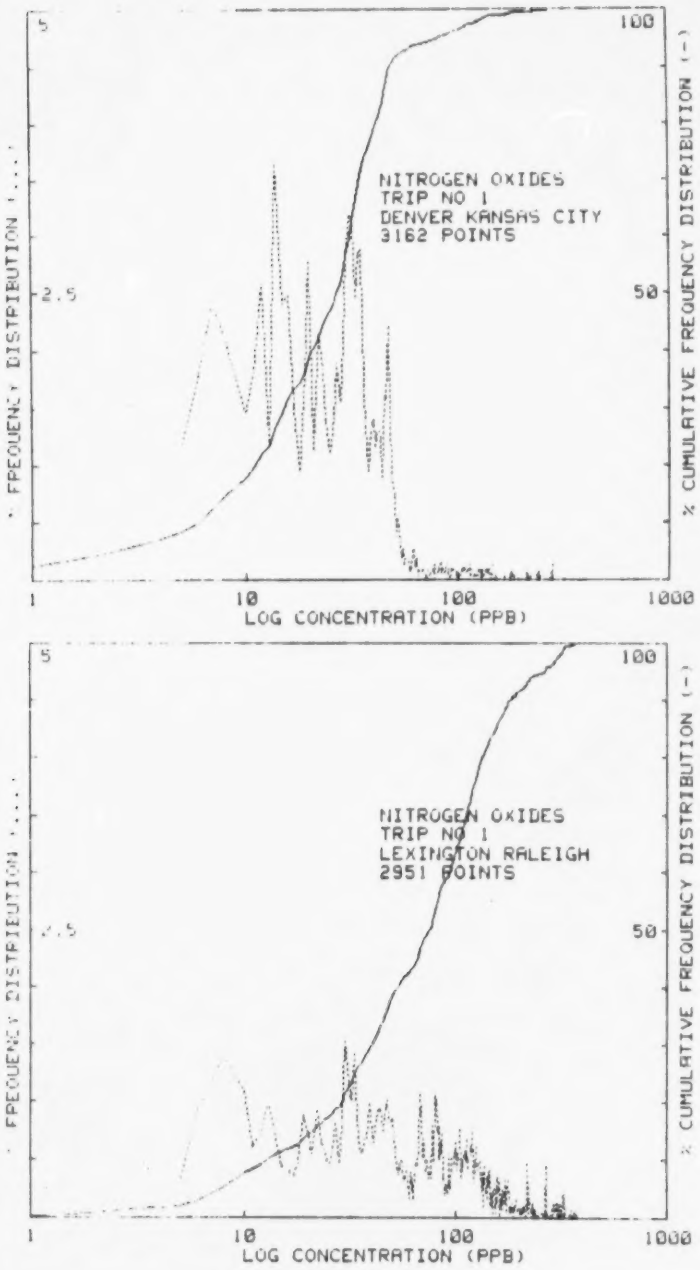
NITROGEN OXIDES
TRIP NO 1
DENVER KANSAS CITY
3162 POINTS
5
2.5
100
50
1
10
100
1000
LOG CONCENTRATION (PPB)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
NITROGEN OXIDES
TRIP NO 1
LEXINGTON RALEIGH
2951 POINTS
5
2.5
100
50
1
10
100
1000
LOG CONCENTRATION (PPB)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)

Figure 45.

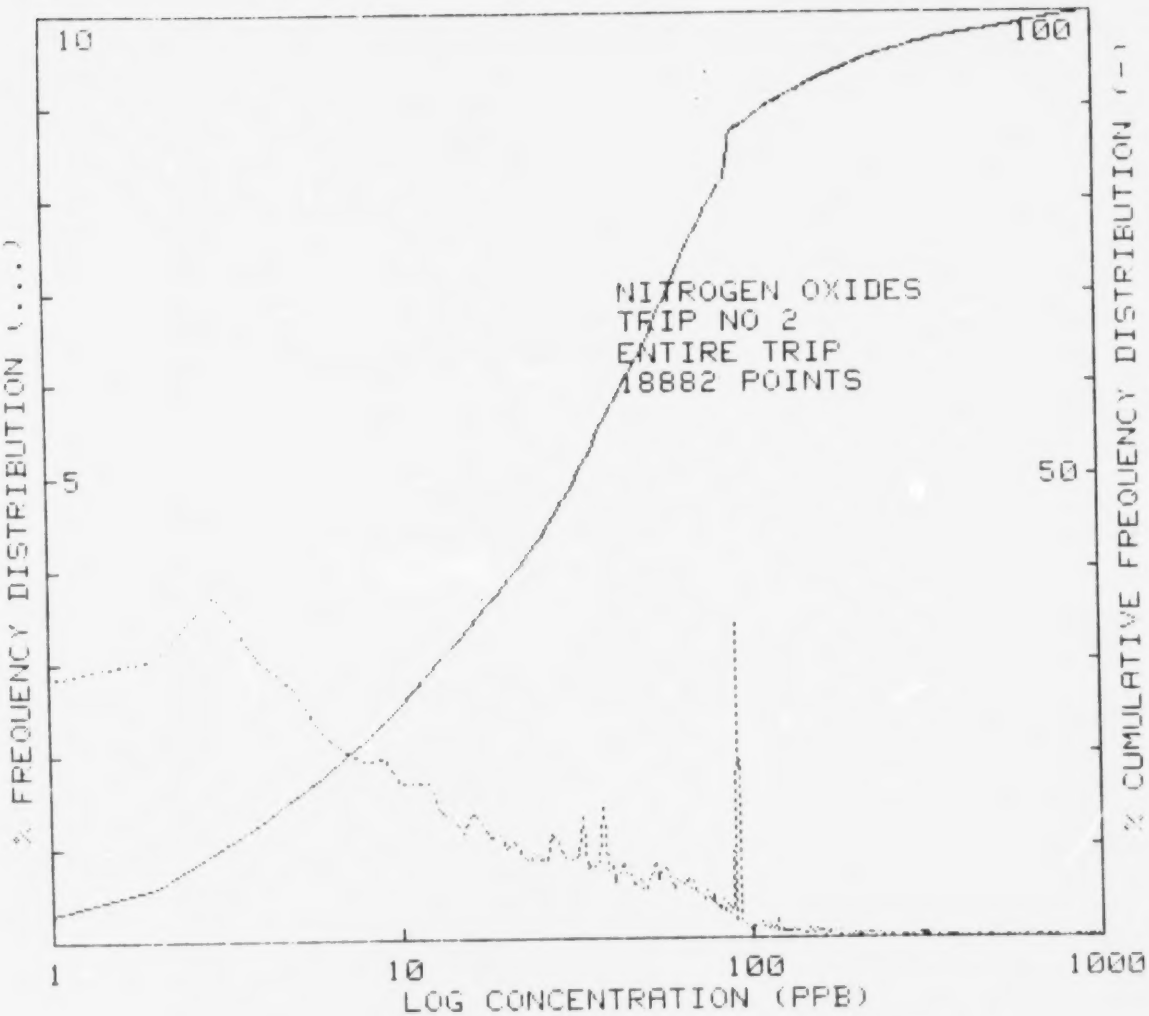
10
100
NITROGEN OXIDES
TRIP NO 2
ENTIRE TRIP
18882 POINTS
5
50
% FREQUENCY DISTRIBUTION (...)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
1
10
100
1000
LOG CONCENTRATION (PPB)

Figure 46.

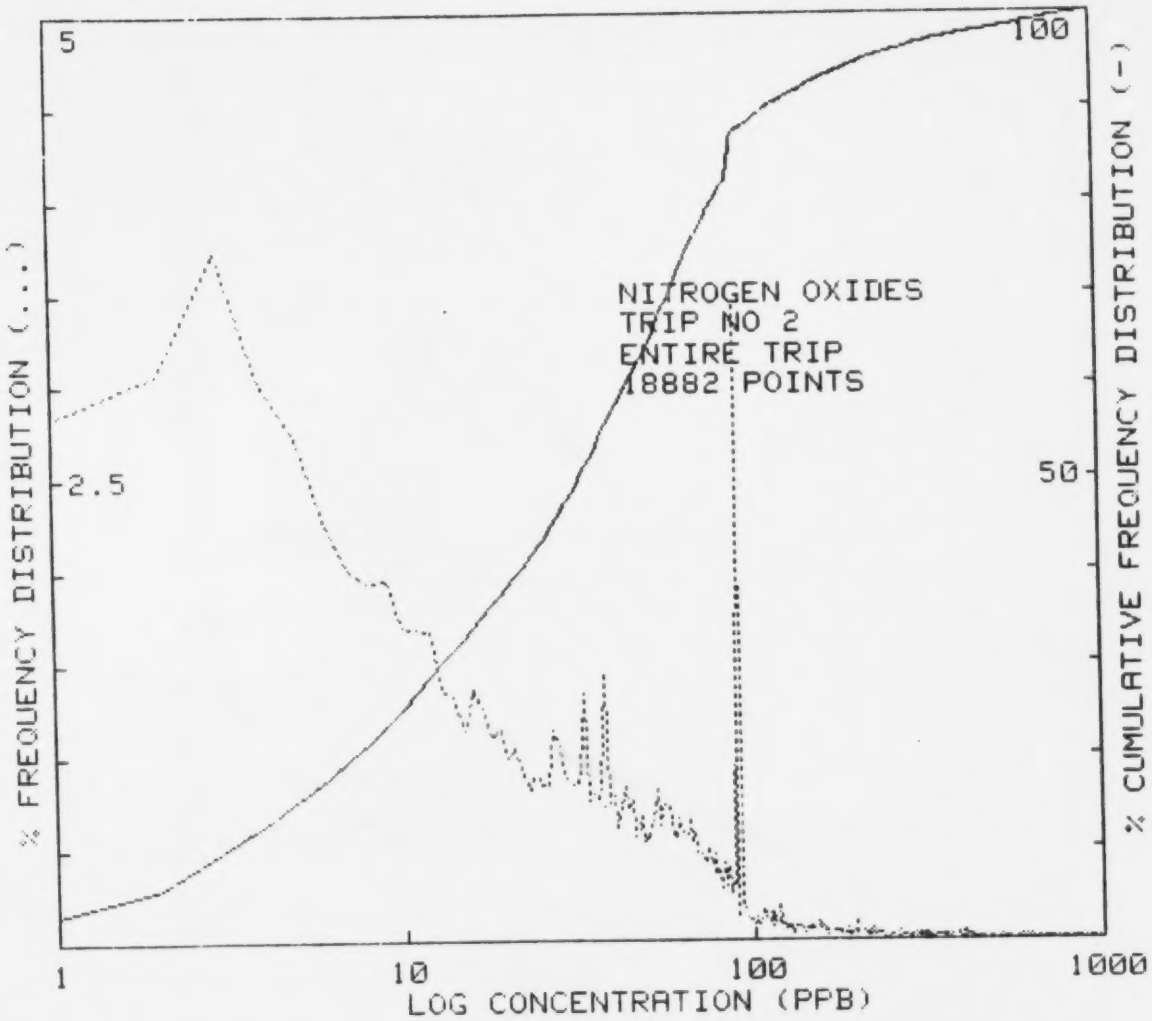
5
2.5
100
50
NITROGEN OXIDES
TRIP NO 2
ENTIRE TRIP
18882 POINTS
% FREQUENCY DISTRIBUTION (...)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
1
10
100
1000
LOG CONCENTRATION (PPB)

Figure 47.

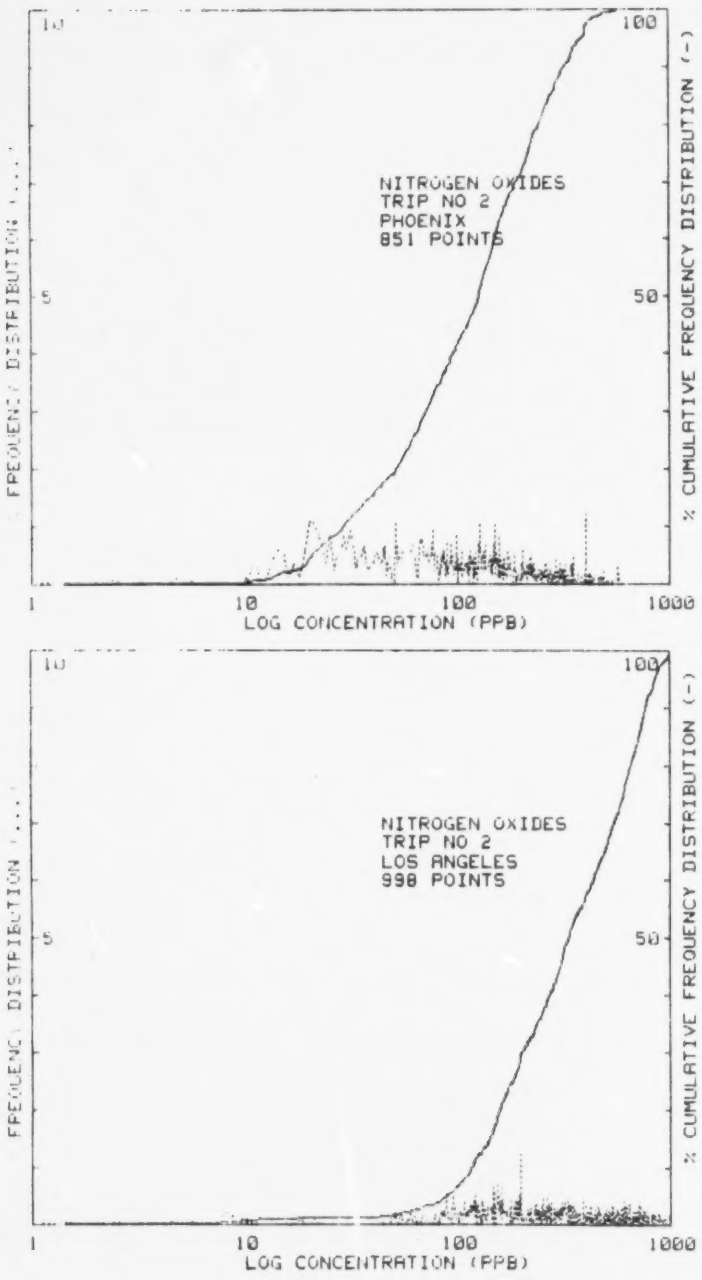
NITROGEN OXIDES
TRIP NO 2
PHOENIX
851 POINTS
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
LOG CONCENTRATION (PPB)
NITROGEN OXIDES
TRIP NO 2
LOS ANGELES
998 POINTS
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
LOG CONCENTRATION (PPB)

Figure 48.

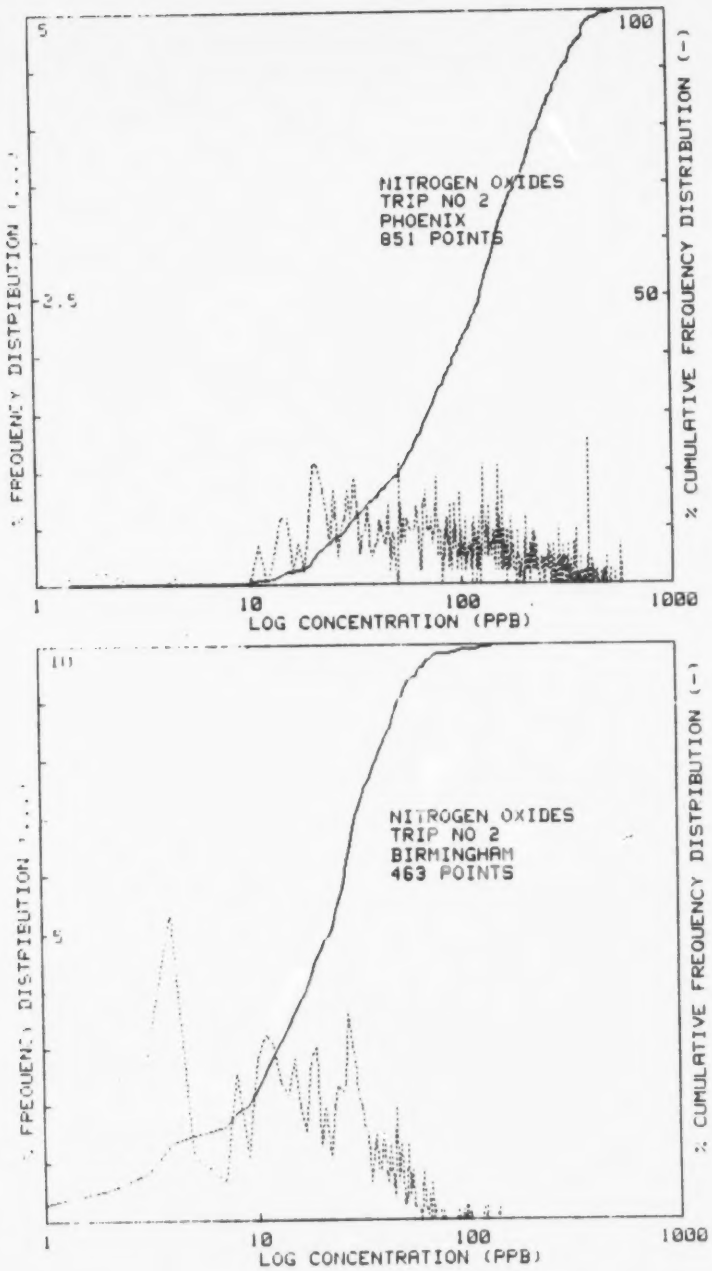
NITROGEN OXIDES
TRIP NO 2
PHOENIX
851 POINTS
% FREQUENCY DISTRIBUTION (-)
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
LOG CONCENTRATION (PPB)
1
10
100
1000
2.5
50
100
NITROGEN OXIDES
TRIP NO 2
BIRMINGHAM
463 POINTS
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
LOG CONCENTRATION (PPB)

Figure 49.

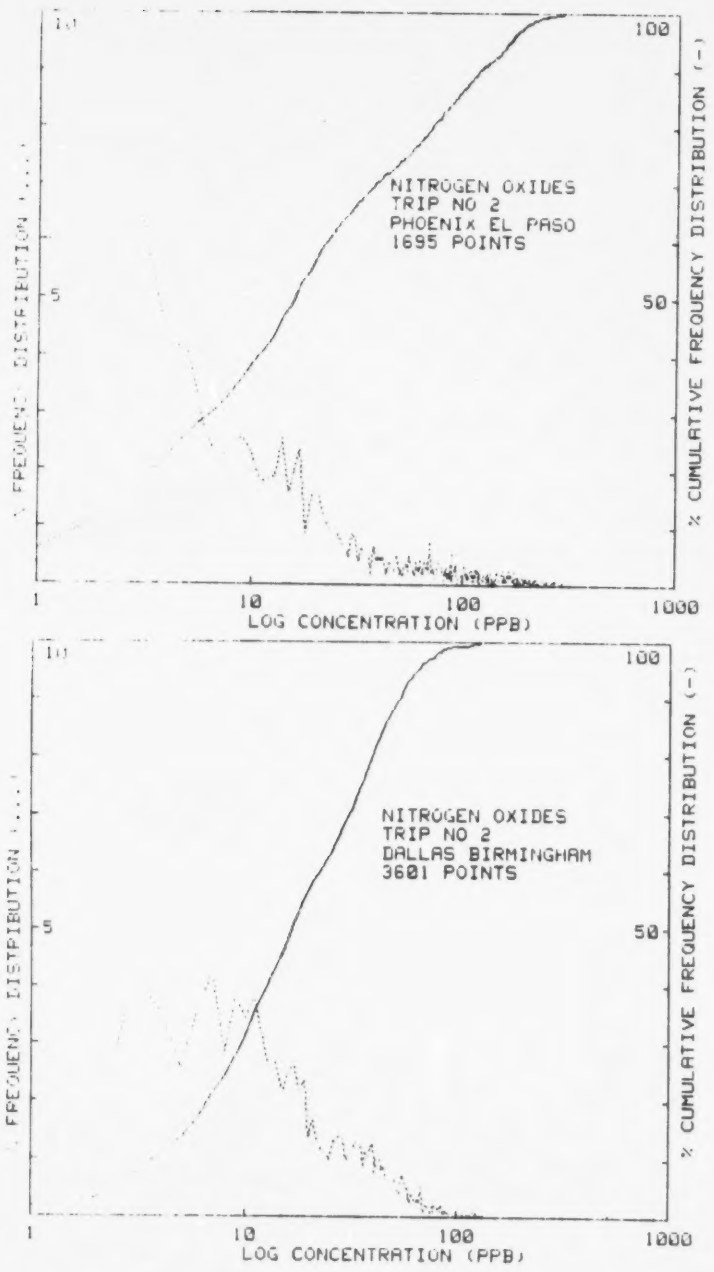
NITROGEN OXIDES
TRIP NO 2
PHOENIX EL PASO
1695 POINTS
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
LOG CONCENTRATION (PPB)
NITROGEN OXIDES
TRIP NO 2
DALLAS BIRMINGHAM
3601 POINTS
% CUMULATIVE FREQUENCY DISTRIBUTION (-)
LOG CONCENTRATION (PPB)

Figure 50.

APPENDIX A

AMMONIA FILTER DATA by ROCKWELL INTERNATIONAL

Atomics International Division
Air Monitoring Center
2421 West Hillcrest Drive
Newbury Park, California 91320
(805) 498-8771

Rockwell International

In reply refer to AMC76-955

14 December 1976

Mr. R. K. Stevens
Environmental Sciences Research Laboratory
Mail Drop 47
Environmental Protection Agency
Research Triangle Park, North Carolina 27711

Dear Bob:

We have analyzed the filters which were returned to us from EMI, Project 148, and summarized the results in the attached table. The right hand column gives the observed concentration of ammonia in the ambient air from each of the two determinations and, below the line, the average of the two results. The first four columns give the information provided by EMI, Inc. on the time, location, and circumstances of the collection of each sample.

As described on pages 21 through 23 of the first Monthly Report under contract 68-02-2463, "Los Angeles Field Modeling and Measurement Study" (LAFMMS), the AMC still has this method under development. Therefore, improvements in the laboratory procedures are still being devised. One evidence of this is that the field blanks in the data reported here had an average of more than 30 times as much ammonium on them as the field blanks from the October LAFMMS sampling. The analysis of two laboratory blanks, which were filters held out of the groups of filters sent to the field, shows that the high blank readings were caused by the laboratory procedure for preparing the filters and not by the shipping and field handling procedures. We are now developing procedures for preparing filters in larger quantities with uniformly low blank ammonium contents.

In three cases, the exposed filters contained less ammonium than the blank, indicating some variability in the blank values. This variability contributes to the scatter in the data. The general range of the results appears reasonable.

We would be very interested to learn how these data compare with those from the chemiluminescent instrument. Should you desire to publish any of these data, we very much hope that their source will be clearly acknowledged.

Sincerely yours,

Willard Richards

Willard Richards
Project Manager

Enclosure

TABLE 3. AMMONIA FILTER DATA

Step Site #	Sample Date/ Time	Sample Location	Comment	μg NH_4/Filter 1 blank	2	3	Ammonia Concentration* $\mu g/m^3$
1	11-8-76 1611-1812	Rt. 81, 4 mi So. of I-10 (vicinity of Lordsburg, NM)	Holder #3 not used.	1.67	2.13	-	0.5
2	11-15-76 1524-1724	Rt. 157, 2 Km No. of Junet, LA. ≈ 10 Km No. of I-20. East of Shreveport, LA.	Low stratus.	2.07	1.58	1.54	-0.6 -0.6 -0.6
3	11-17-76 1135-1335	Miss. 49, 8 Km No.Pelahatchee, MS. 30 Km East of Jackson, MS.	Used filter batch 1508, received Jackson, MS.	1.23	0.98	1.41	-0.3 +0.3 0.0
4	11-18-76 1425 EST 1525	9 Km S.Ragland, AL. ≈ 60 Km ENE Birmingham. Refuge Church Yard.	Pine forest area. Filter batch 1508, low wind speed. Clear & warm (70°F)	0.41	1.95	1.47	1.8 1.3 1.5
5	11-19-76 1342 EST 1542	12 Km NW Conmerce, GA (near a creek) btw. Homer & Maysville on Rt. 98, GA.	Filter batch 1499 (received Birmingham, AL.)	0.45	1.23	1.13	0.9 0.8 0.8
6	11-20-76 1410 EST 1610	25 Km WSW Chapel Hill, NC. 18 Km NE Silver City, NC.	Filter batch 1499 Near dairy farm/ feedlot, heavy agricultural region. Hay odor. NH_3 denuder removes much of NOx signal. Also in SOx plume.	0.58	2.80	1.69	2.6 1.3 1.9
Laboratory blanks				0.54 1.81			

*The ammonia concentration from each filter and the mean is given.

The technique uses oxalic acid coated on a glass fiber filter. Ambient air is drawn through a 1 μm fluoropore filter to remove all particulates, then through the glass fiber filter coated with oxalic acid. The ammonia gas passes through the first filter and is collected on the second. The filters are subsequently analyzed.